California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Modeling Analyses of Data Captured During the CRPAQS Field Program

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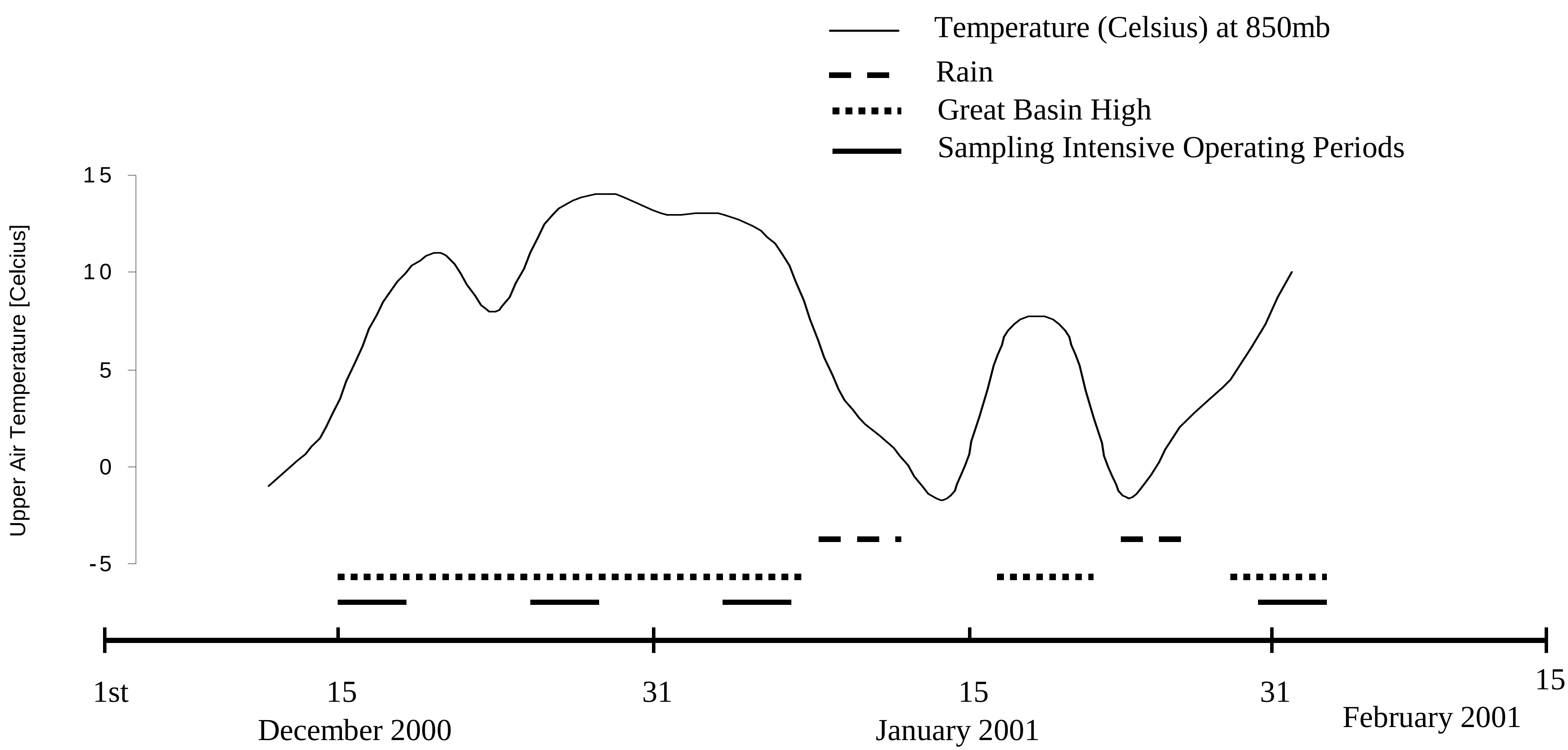
Acknowledgements

- Karen Magliano, Ajith Kaduwela, Vernon Hughes
- Paul Allen, Paul Livingstone
- Everyone who made measurements during CRPAQS
- Zhan Zhao, Shuhua Chen

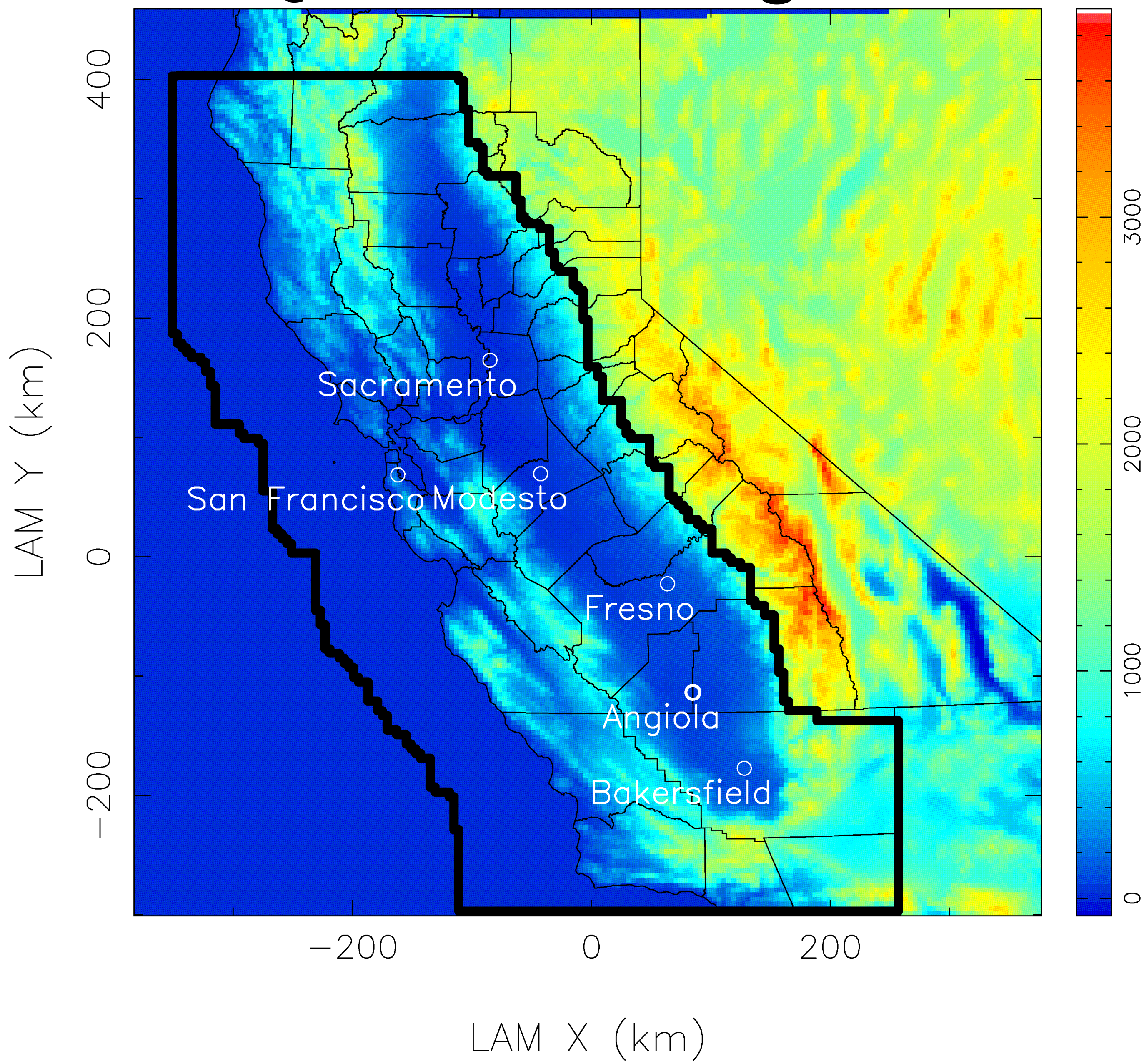
- San Joaquin Valleywide Air Pollution Study Agency
- United States Environmental Protection Agency

Meteorological Patterns During CRPAQS Winter Intensive



Source: J. Herner, J. Aw, O. Gao, D.P. Chang, and M.J. Kleeman, "Size and Composition Distribution of Airborne Particulate Matter in Northern California: 1 – Particulate Mass, Carbon, and Water-Soluble Ions", J. Air. Waste Manag. Assoc., 55: 30-51, 2005.

CRPAQS Modeling Domain



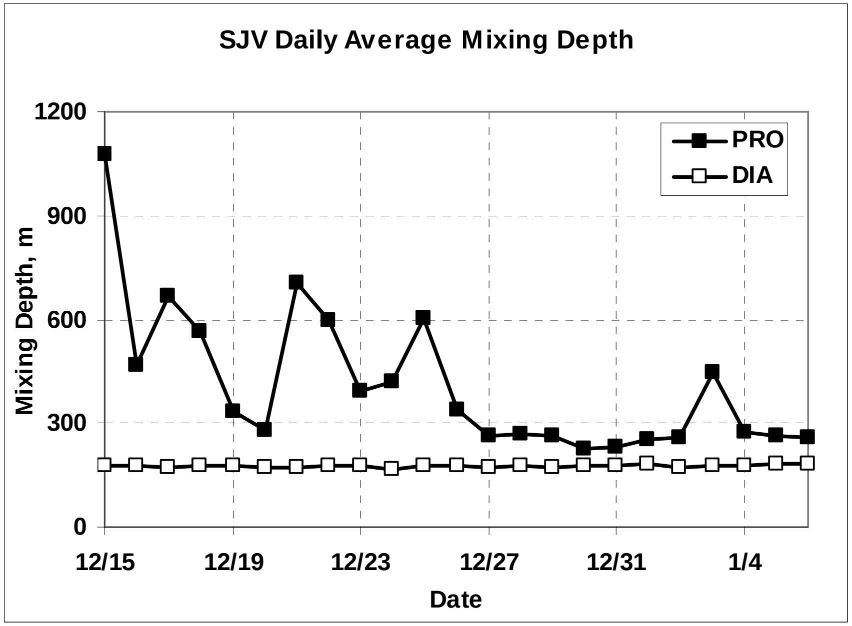
Phase 1 Project Objectives

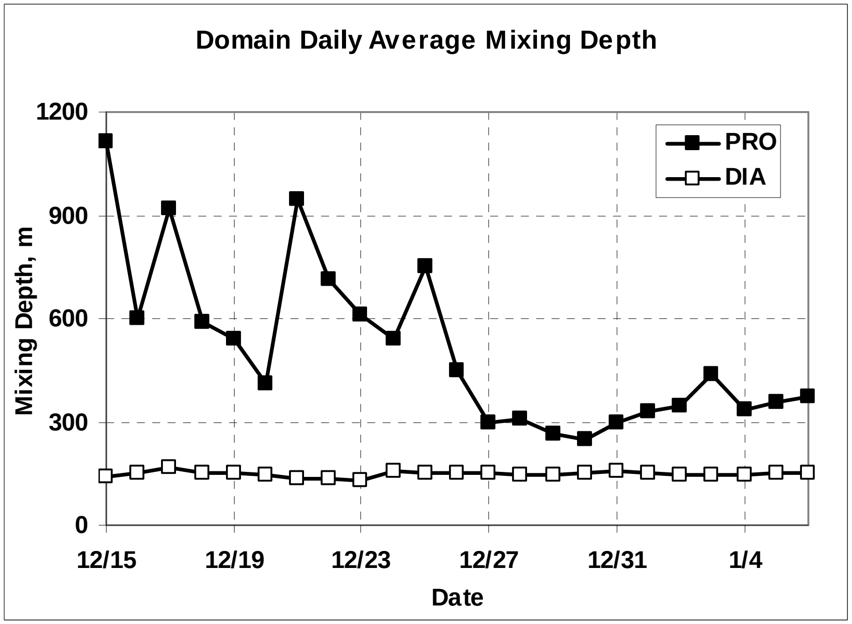
1. Create a speciated emissions inventory for air quality modeling (complete)
2. Generate diagnostic meteorology fields and compare to prognostic fields (complete)
3. Evaluate sub-grid emissions transformation processes (complete)
4. Generate diagnostic air quality fields for initial conditions and boundary conditions (complete)

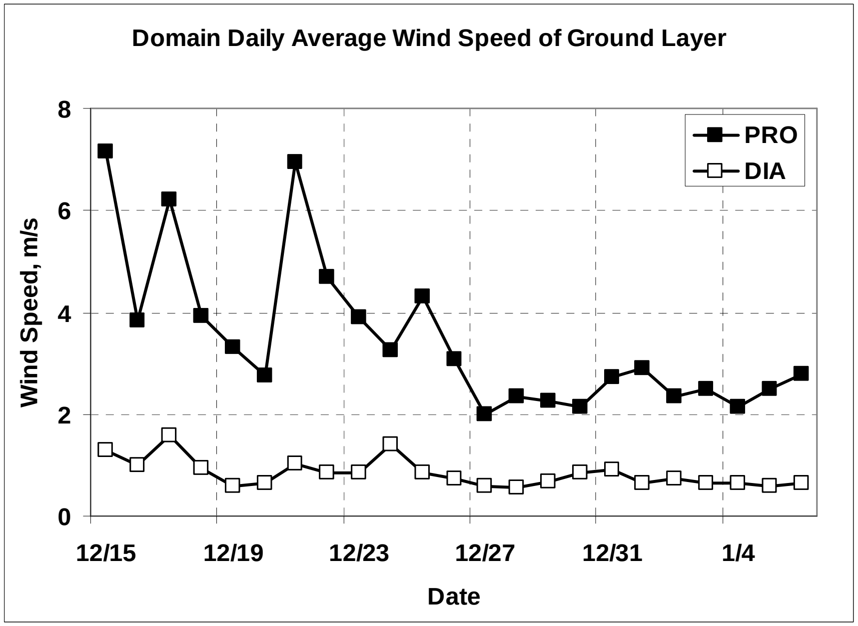
Phase 1 Project Objectives

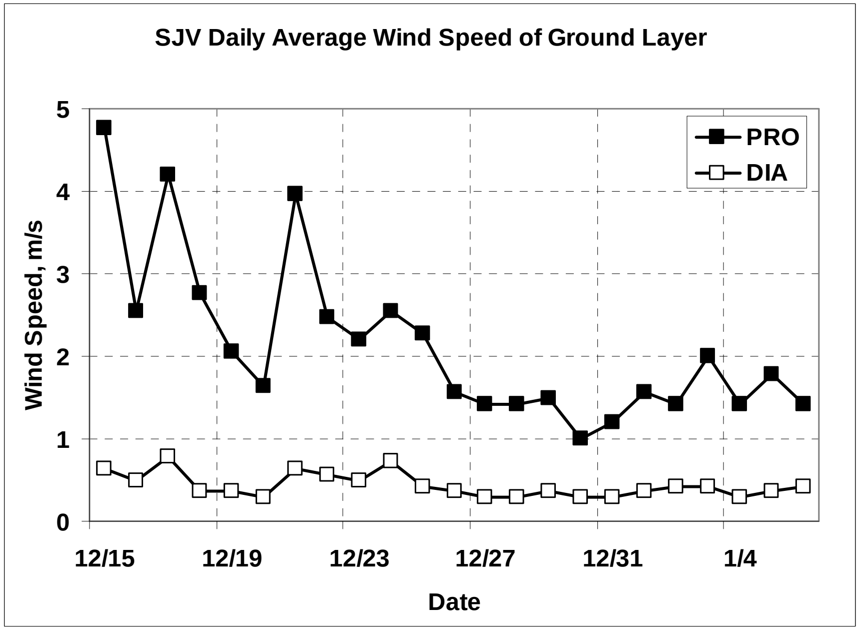
1. Modify UCD/CIT model to simulate larger CRPAQS domain and incorporate faster thermodynamic calculations (complete)
2. Perform calculations for CRPAQS IOPs 1-3 and validate basecase performance (complete)
3. Source Apportionment of Primary PM (complete)
4. Source Apportionment of Secondary PM (complete)
5. Regional Impacts on PM (complete)

Diagnostic vs. Prognostic Meteorology

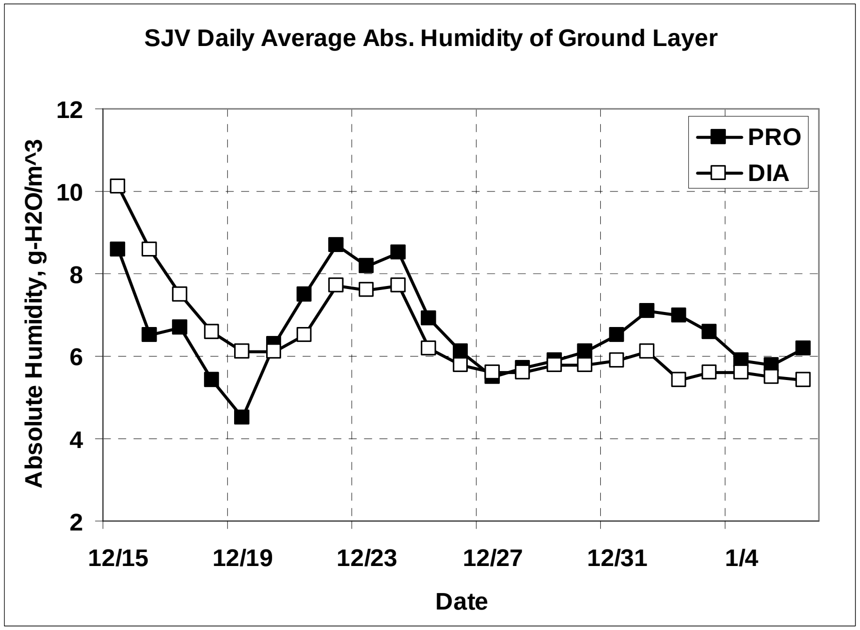


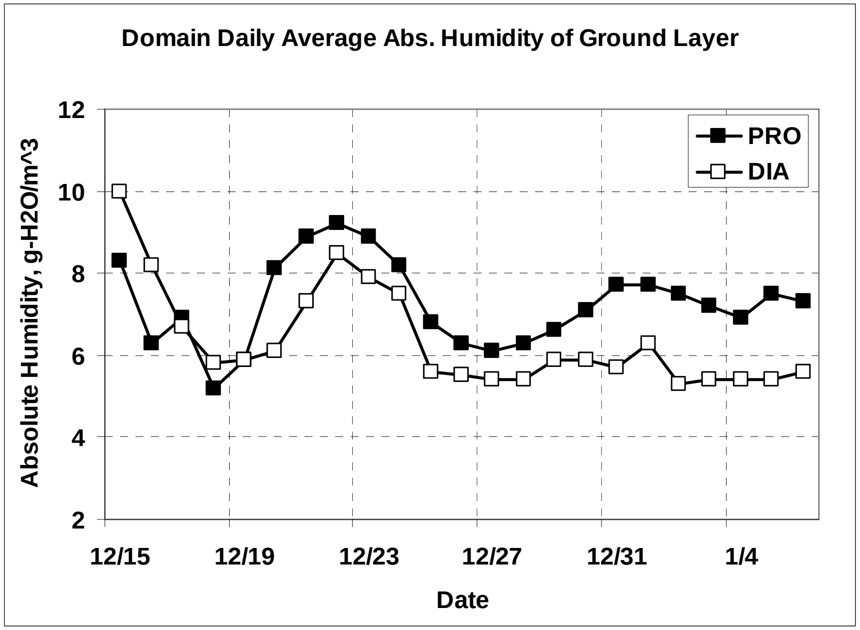


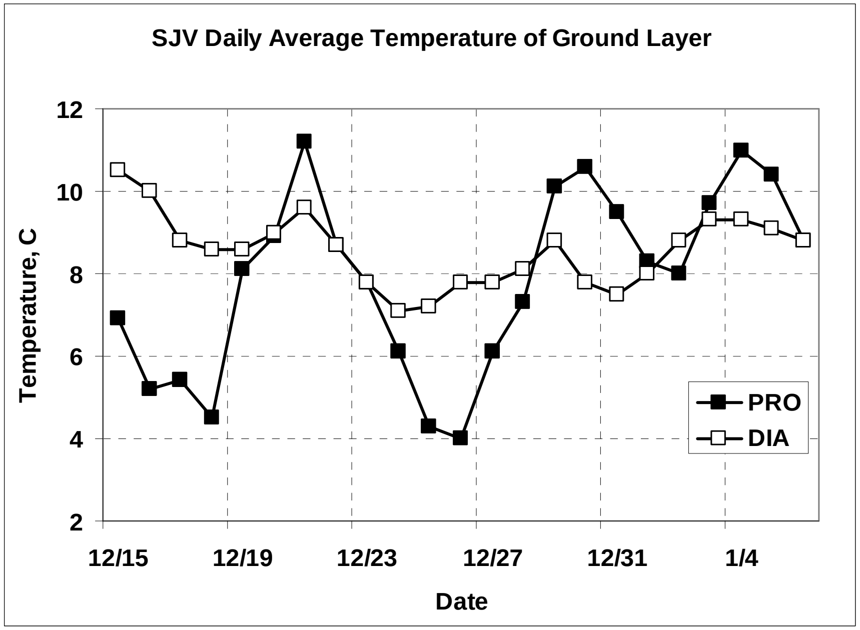


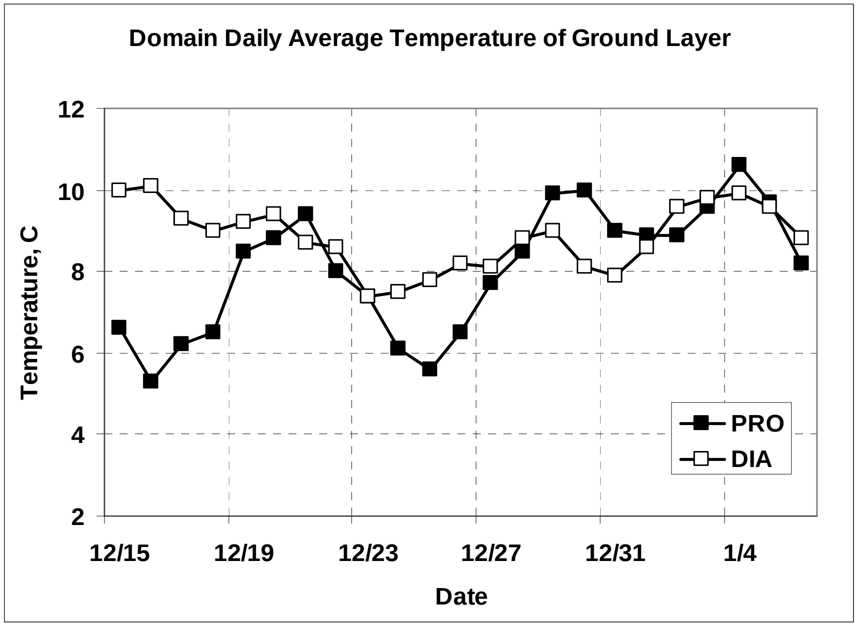


Diagnostic vs. Prognostic Meteorology

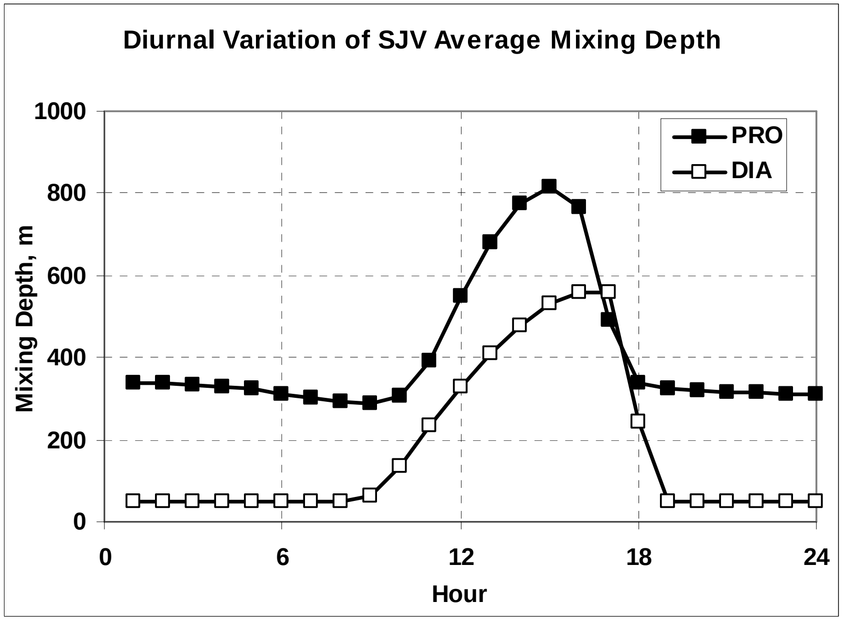


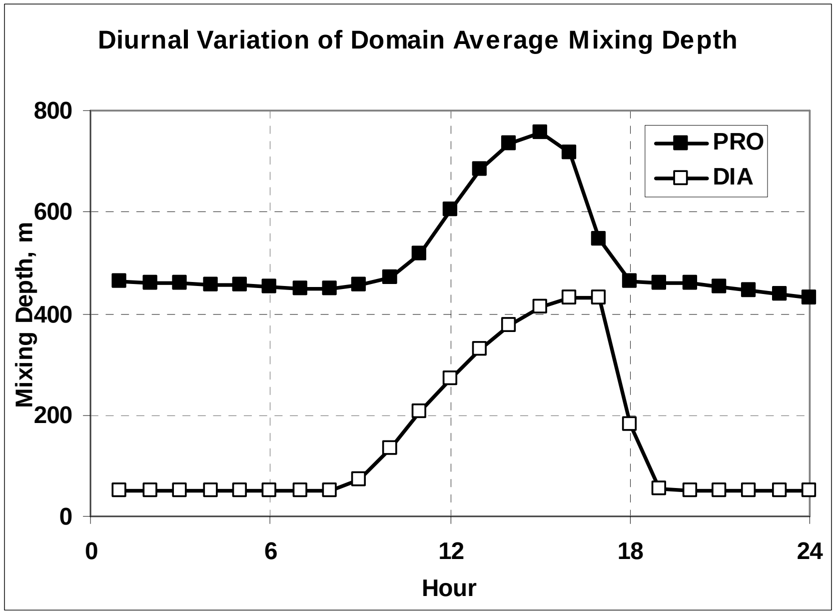


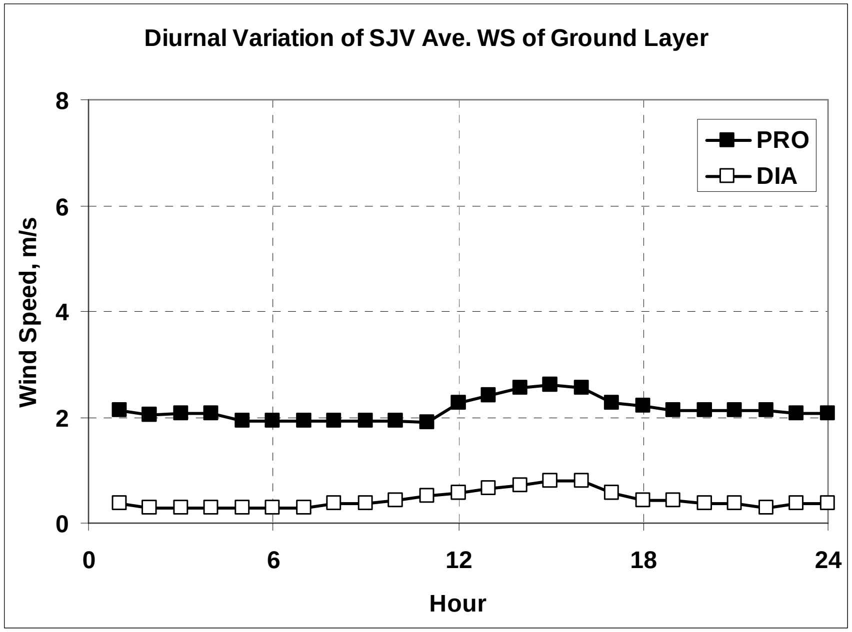


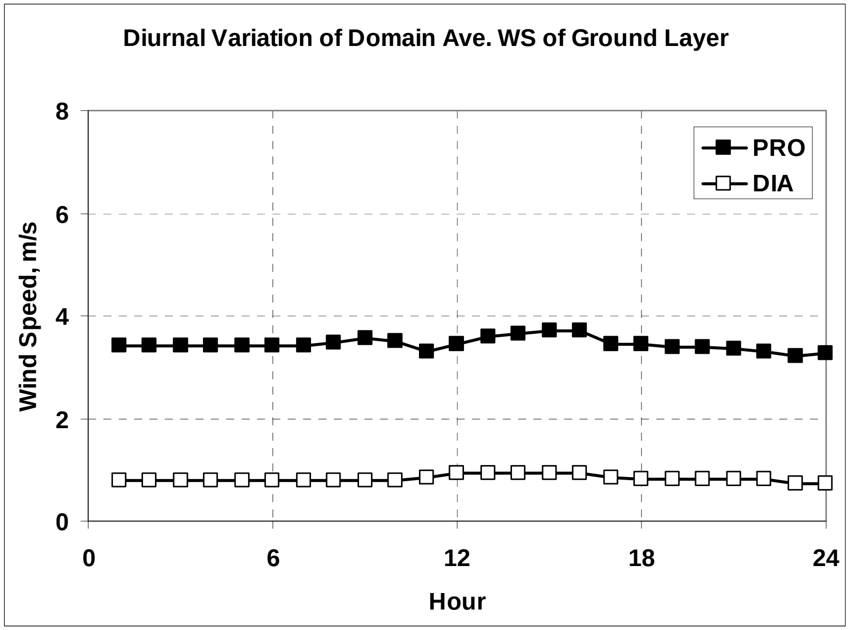


Diagnostic vs. Prognostic Meteorology

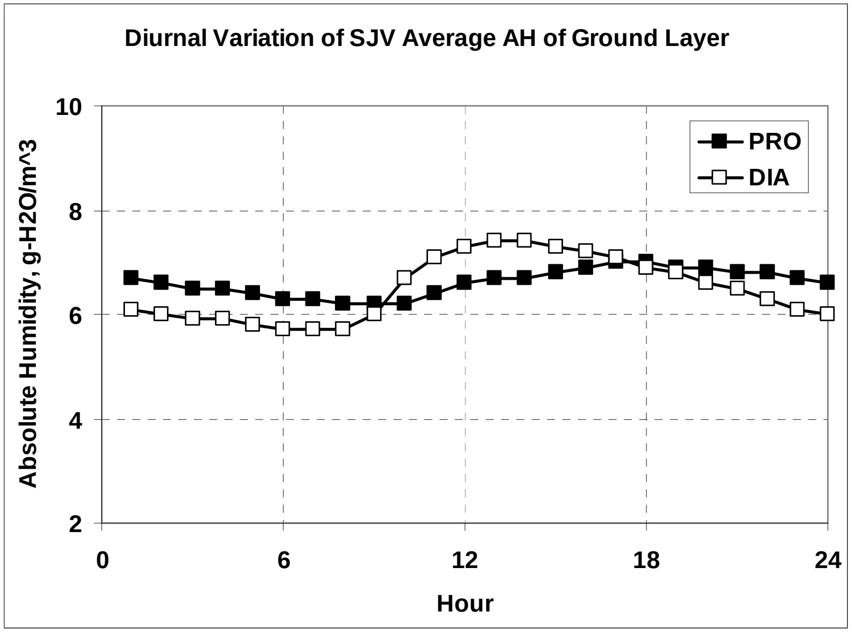


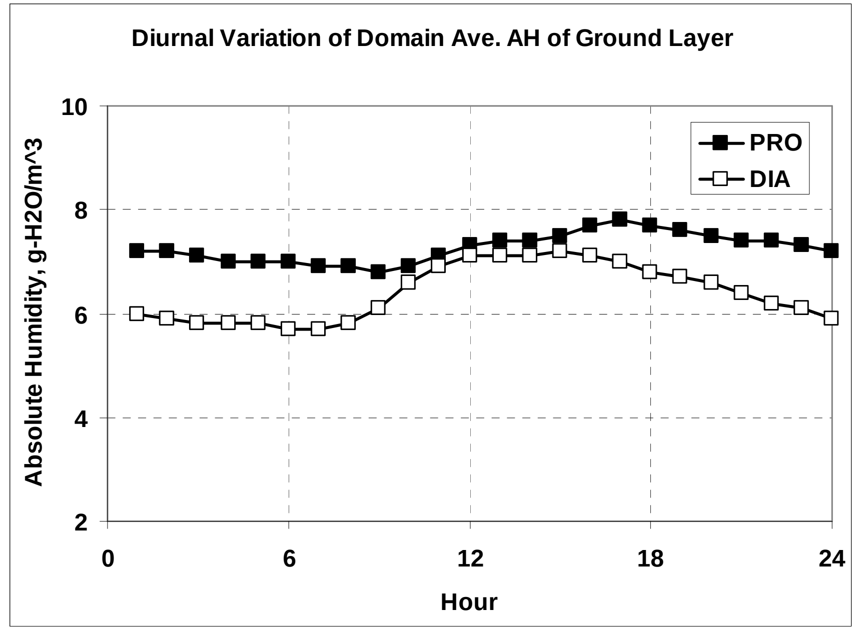


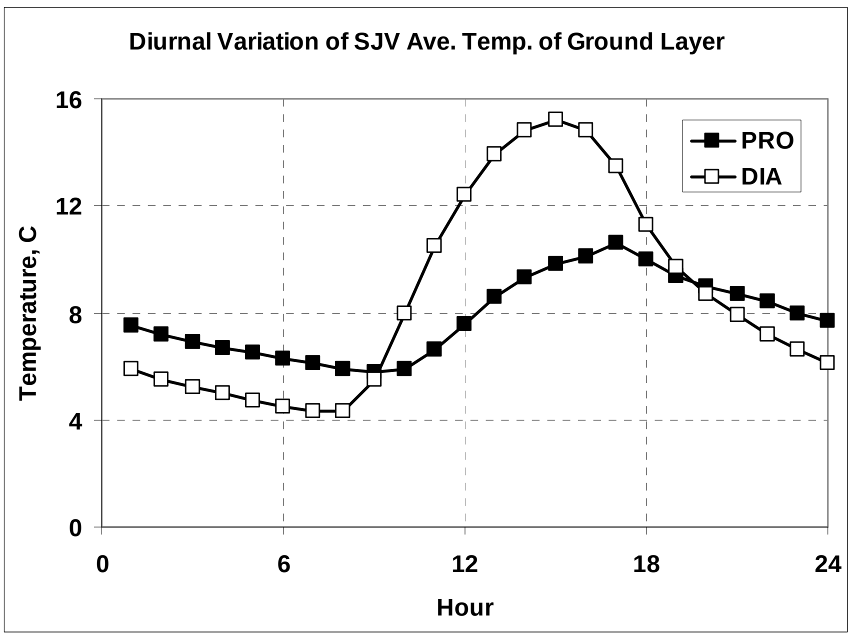


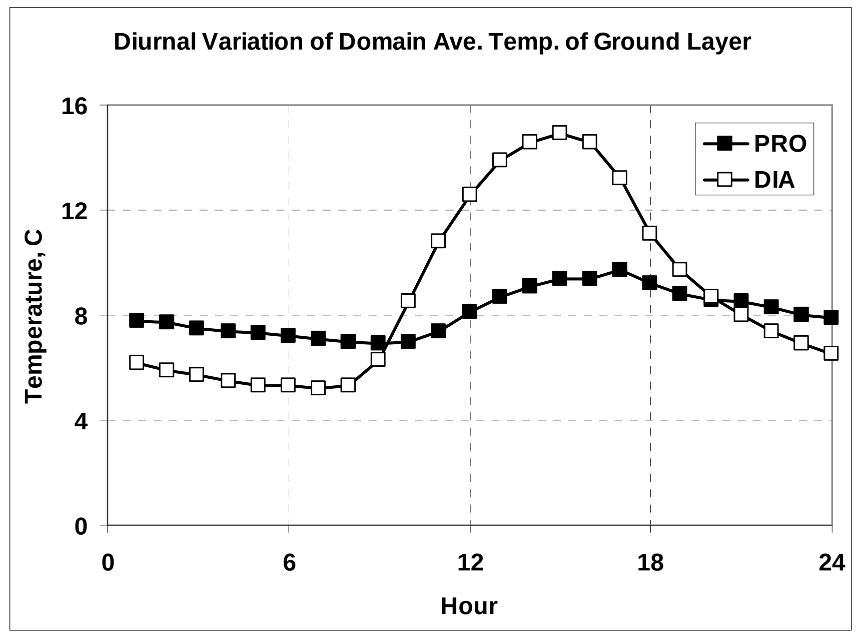


Diagnostic vs. Prognostic Meteorology

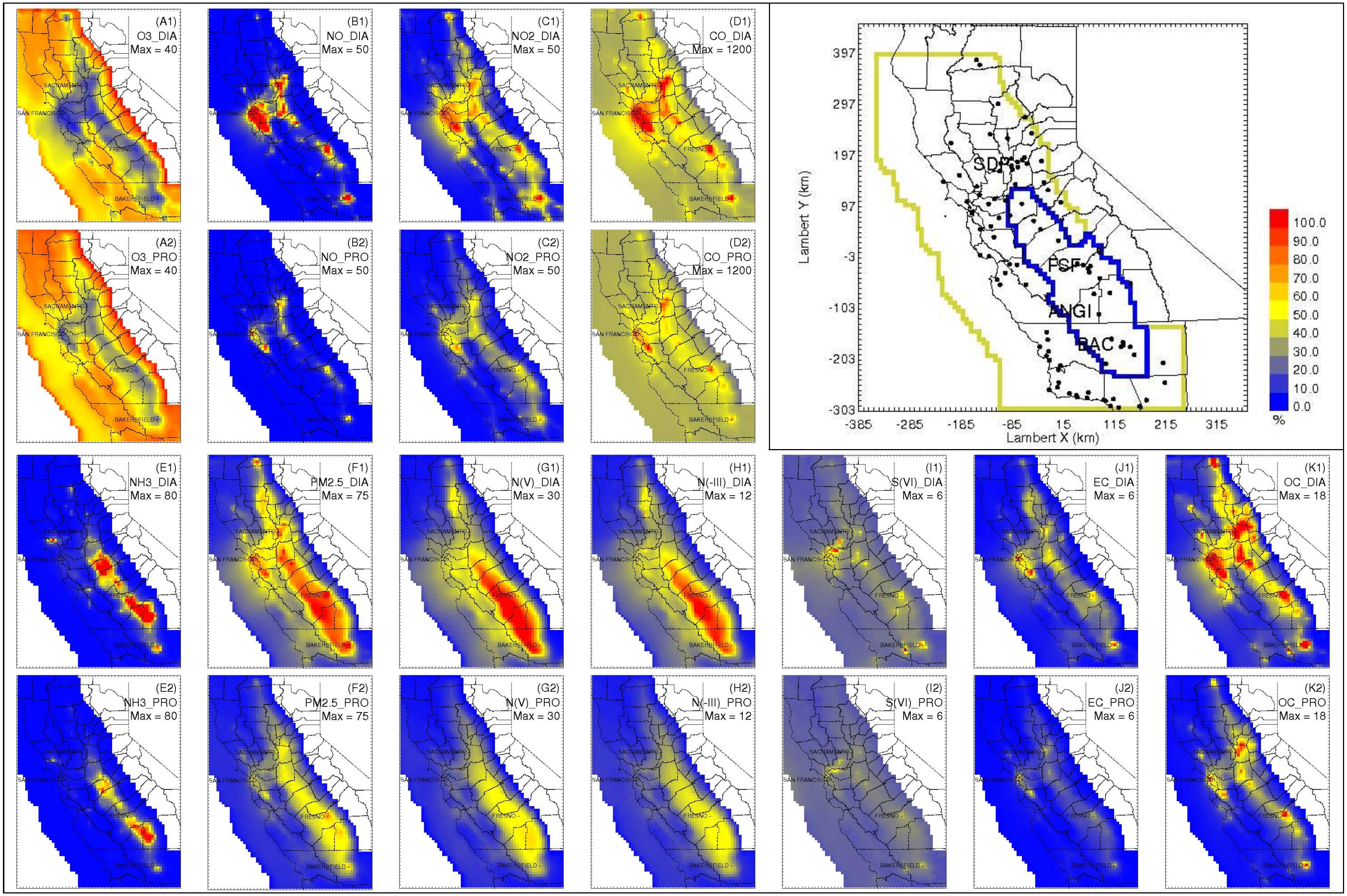








Diagnostic vs. Prognostic Results



Diagnostic vs. Prognostic Conclusions

- Similar results, but higher mixing depths and larger surface wind speeds dilute prognostic results
- Diagnostic results are slightly more accurate at the surface

CO

Black Line – measurements

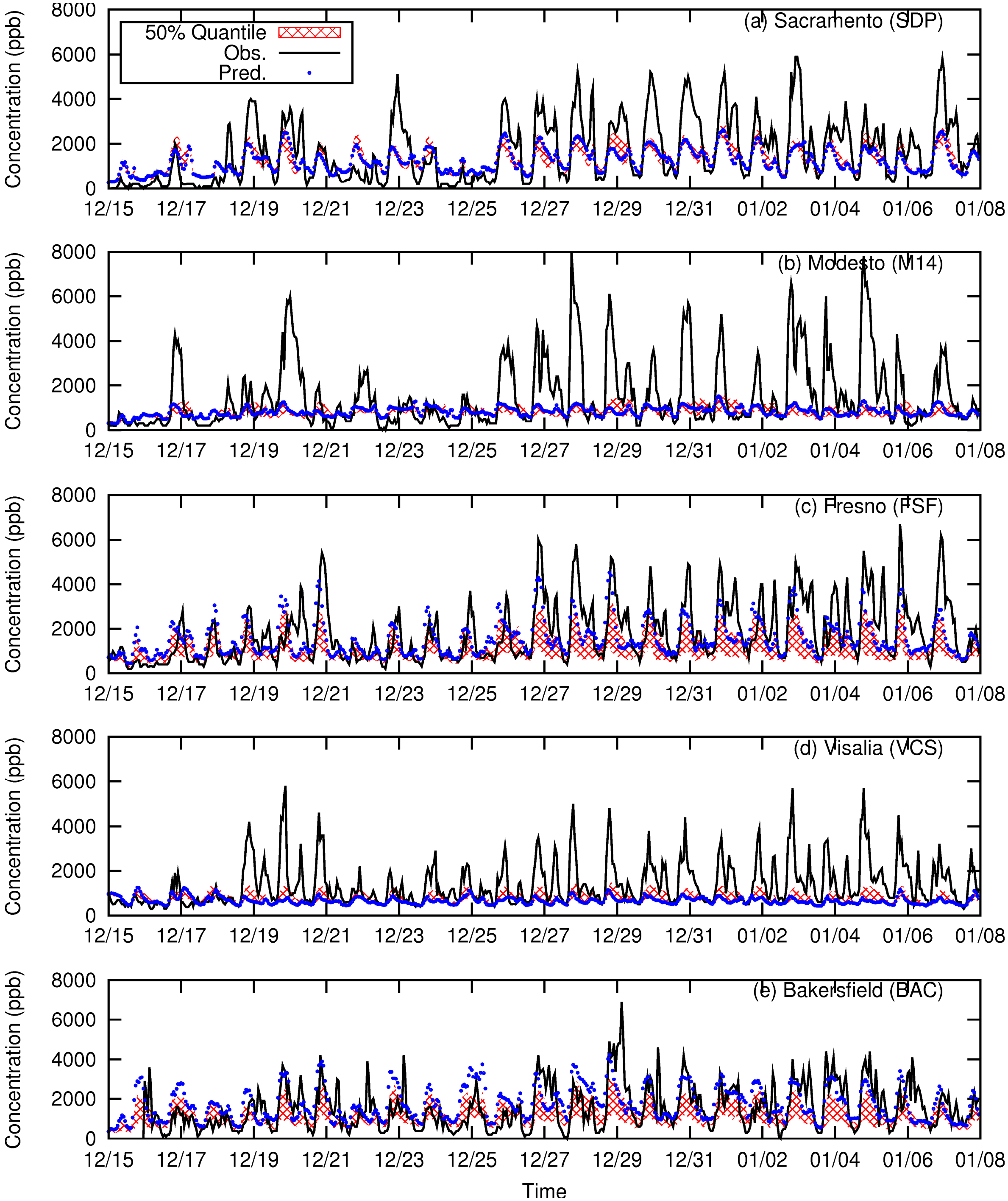
Blue Line – predictions

Red Shading – Mid 50% Quantile within 10km of monitor

Sacramento, Fresno, Bakersfield have correct diurnal pattern

Modesto and Visalia do not reproduce observed diurnal pattern

Source: Q. Ying, J. Lu, P. Allen, P. Livingstone, A. Kaduwela, and M. Kleeman “Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part I. Base Case Model Results.”, Atmos. Env., in press, 2008.

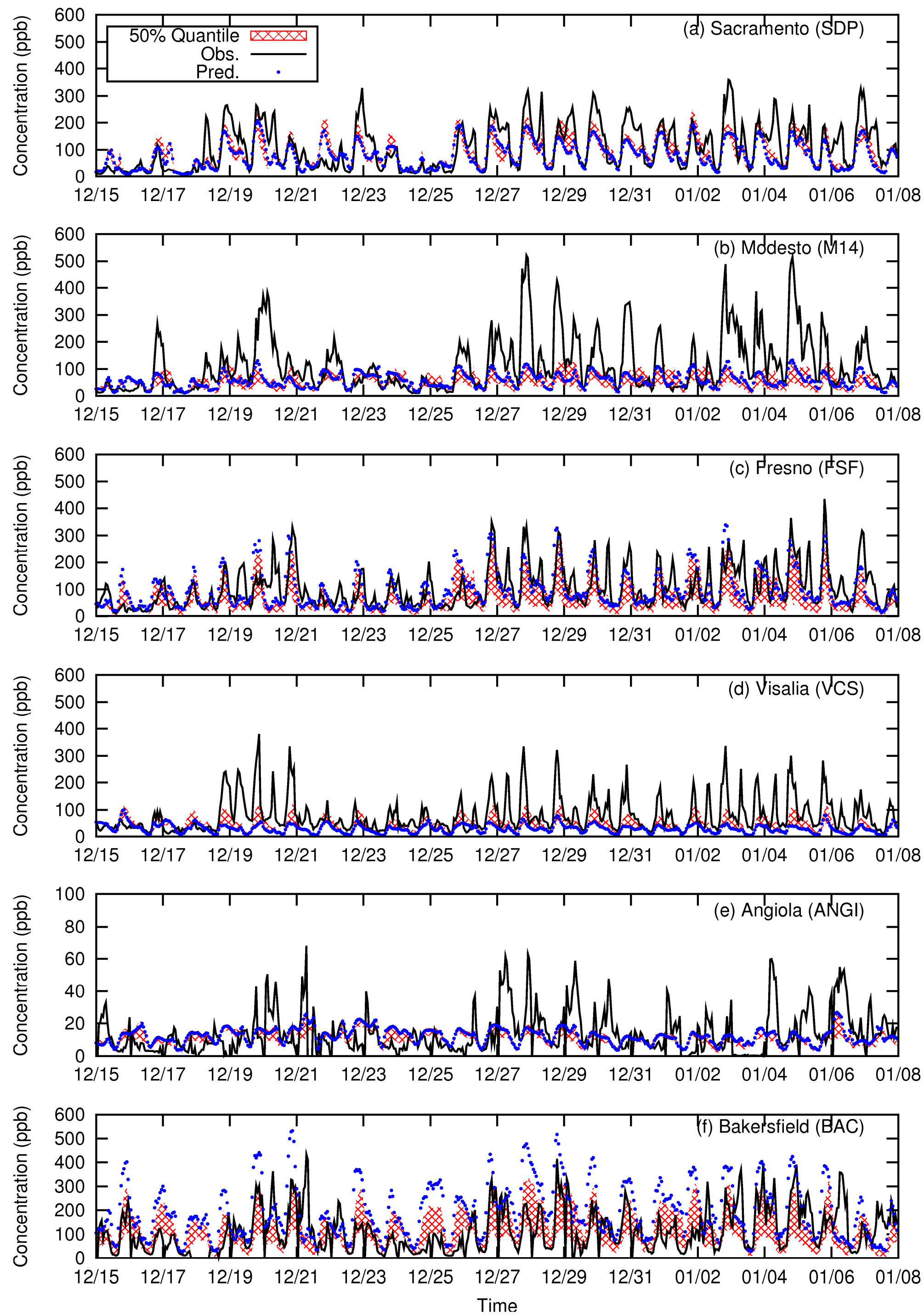


NOx

Good agreement at Sacramento and Fresno

Slight over-prediction at Bakersfield, but spatial gradients are sharp

Miss the diurnal trend at Modesto, Visalia, and Angiola (what sources drive the observed trends?)



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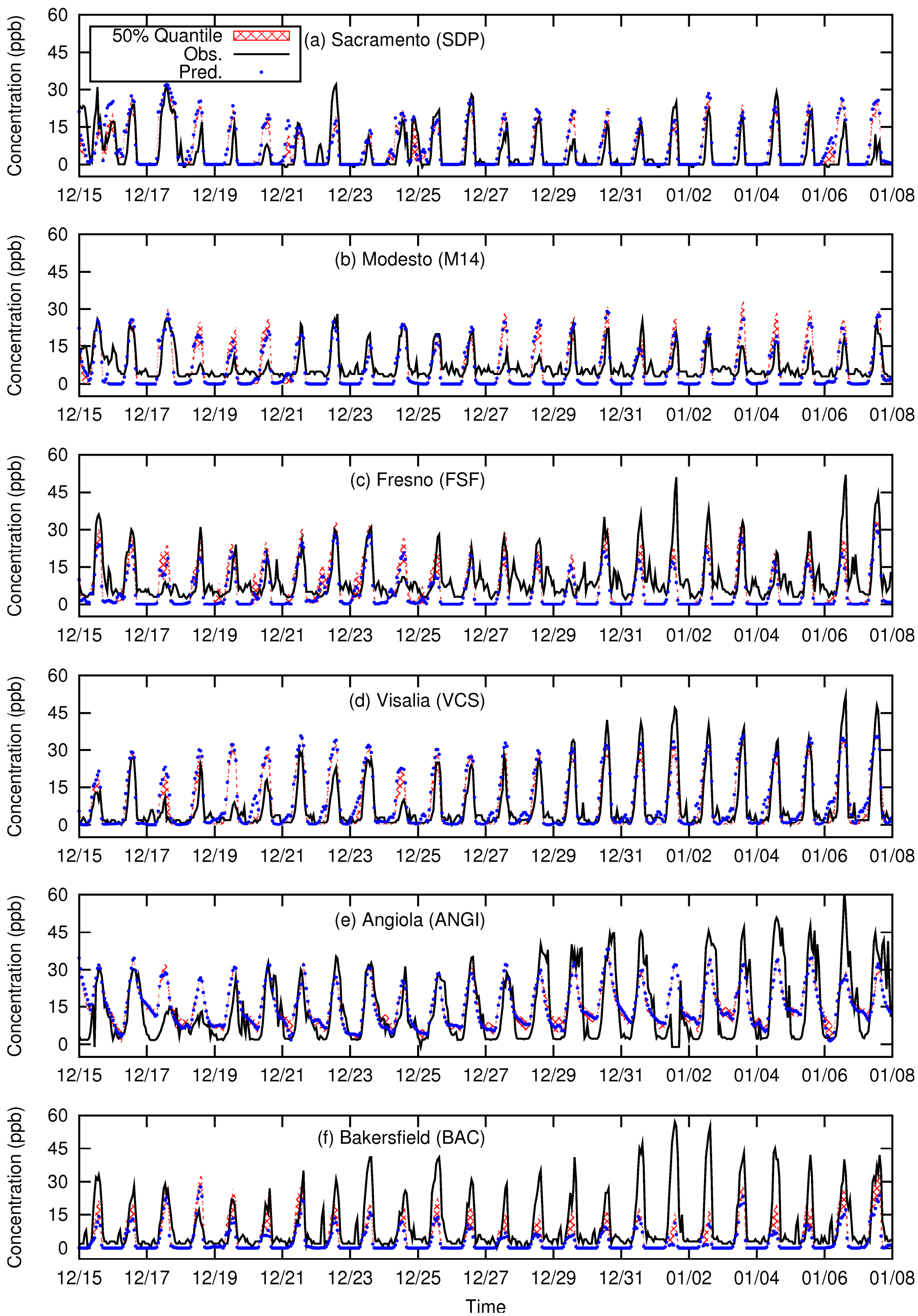
O3

Good agreement at most stations

Majority of the ozone is background that mixes down to the surface during the middle of the day

Background O3 is actually the dominant oxidant in the atmosphere



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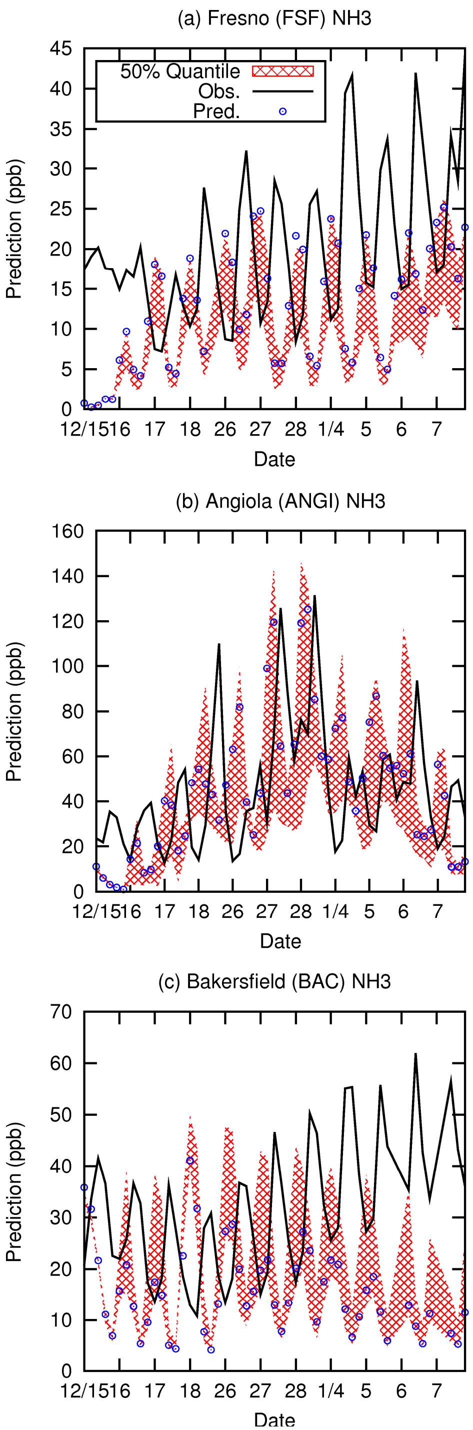
NH3

Overall concentrations are order-of-magnitude correct

Diurnal cycle is exactly opposite measured values

Nitrate production is limited by production of nitric acid, so the details are not critical



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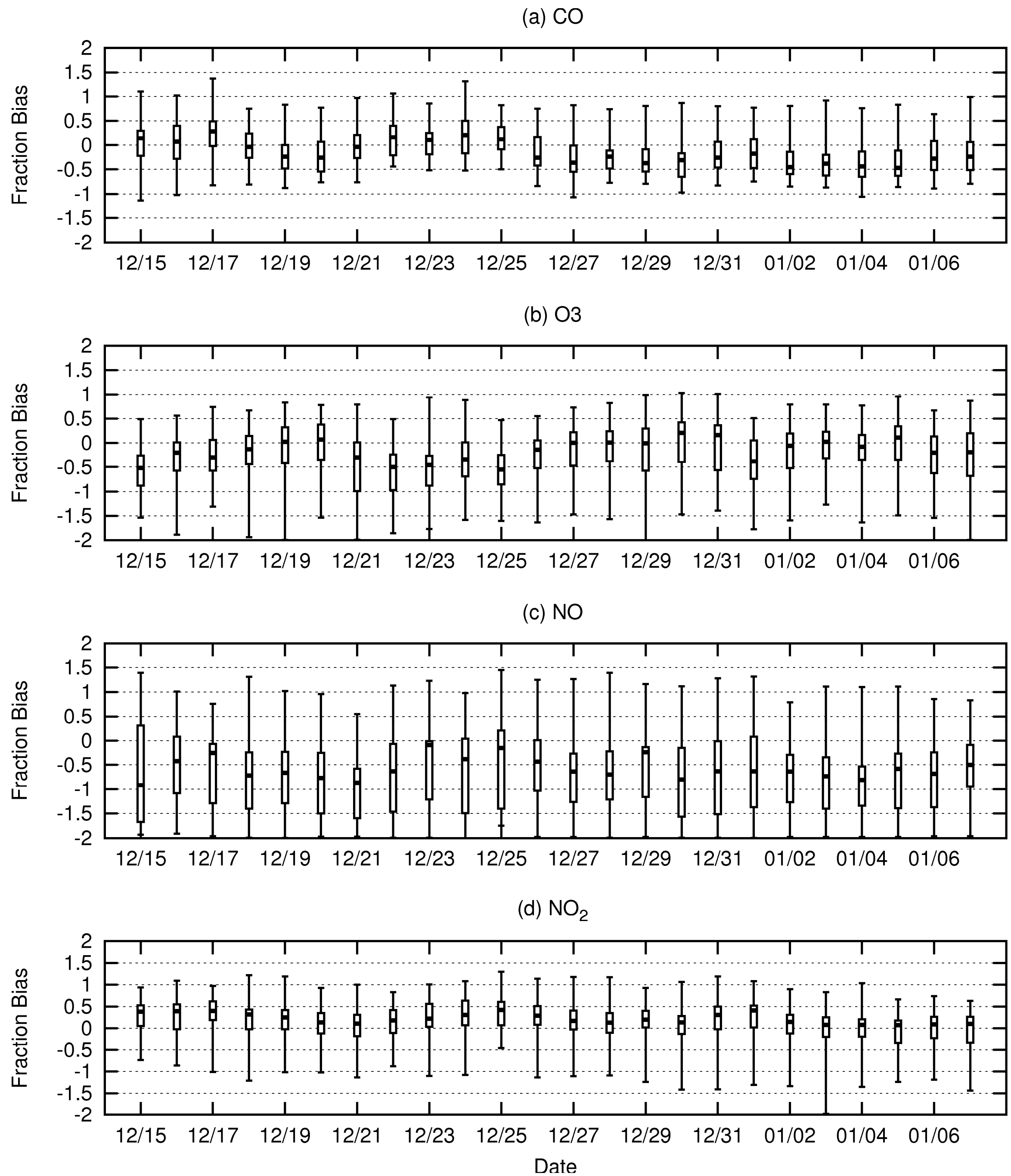
Fractional Bias at All Stations

$$FB = \frac{2}{N} \times \sum \frac{C_{p,i} - C_{o,i}}{C_{p,i} + C_{o,i}}$$



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PM2.5 Mass

Black Line – measurements

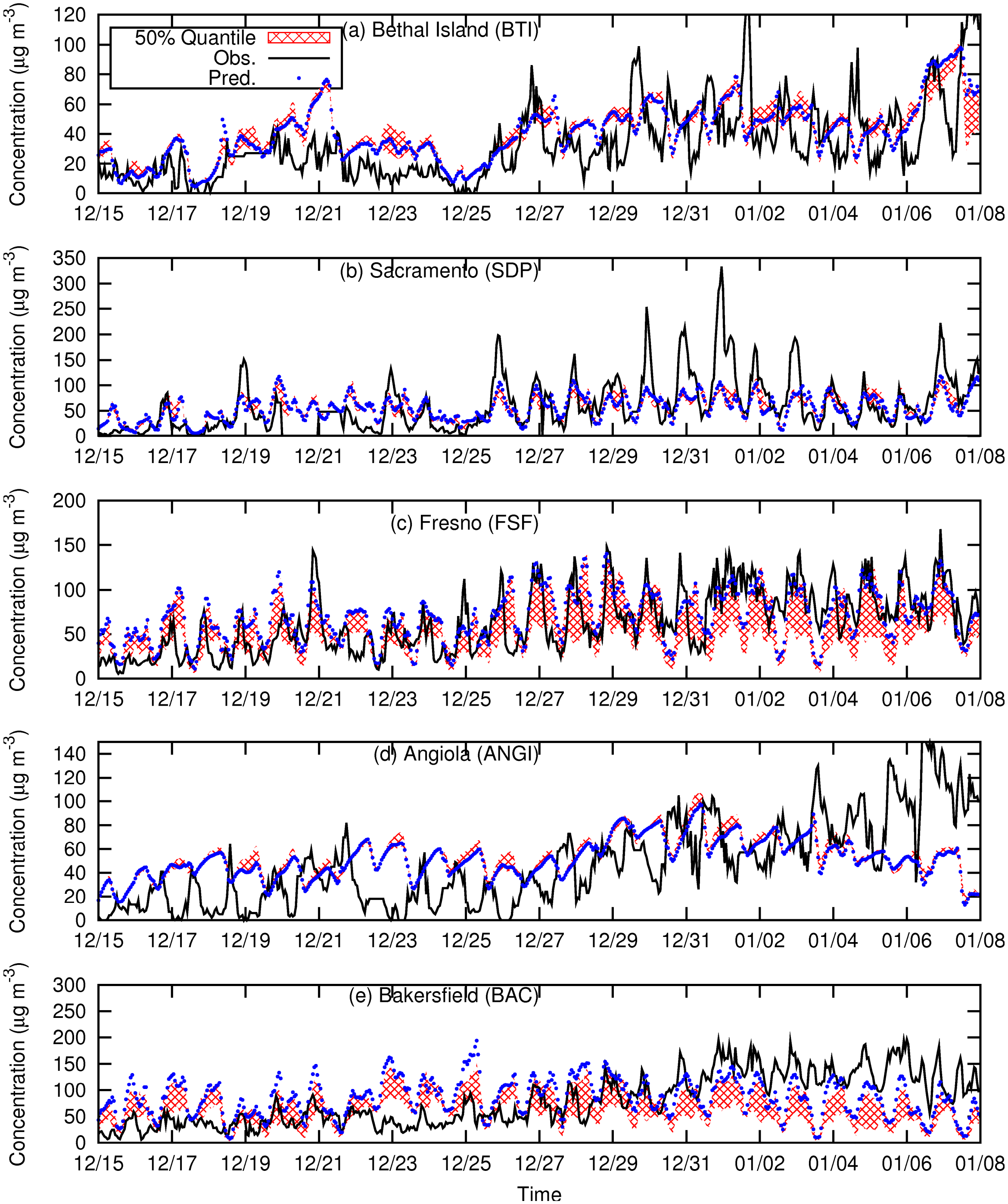
Blue Line – predictions

Red Shading – Mid 50% Quantile within 10km of monitor

Major trends are captured at most stations

Under-prediction of mass at Angiola and Bakersfield near the end of the episode

Source: Q. Ying, J. Lu, P. Allen, P. Livingstone, A. Kaduwela, and M. Kleeman "Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part I. Base Case Model Results.", Atmos. Env., in press, 2008.



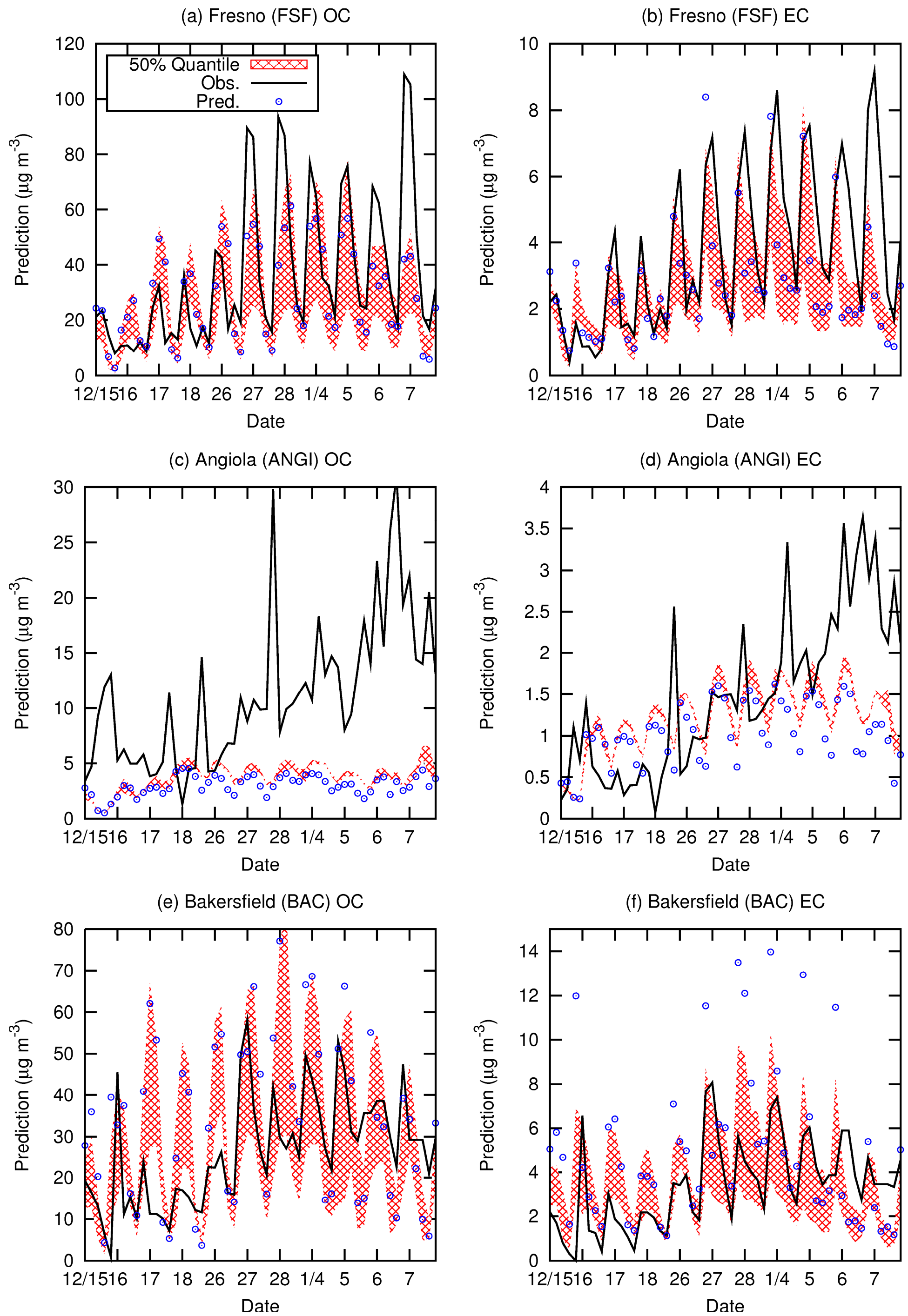
PM2.5 OC + EC

Black lines = measurements
Blue circles = predictions
Red Shading – Mid 50% Quantile within 10km of monitor

Diurnal pattern predicted correctly at urban sites

Peak values show reasonable agreement, especially considering the sharp gradients

Rural Angiola predictions are low. Where is the EC+OC coming from?



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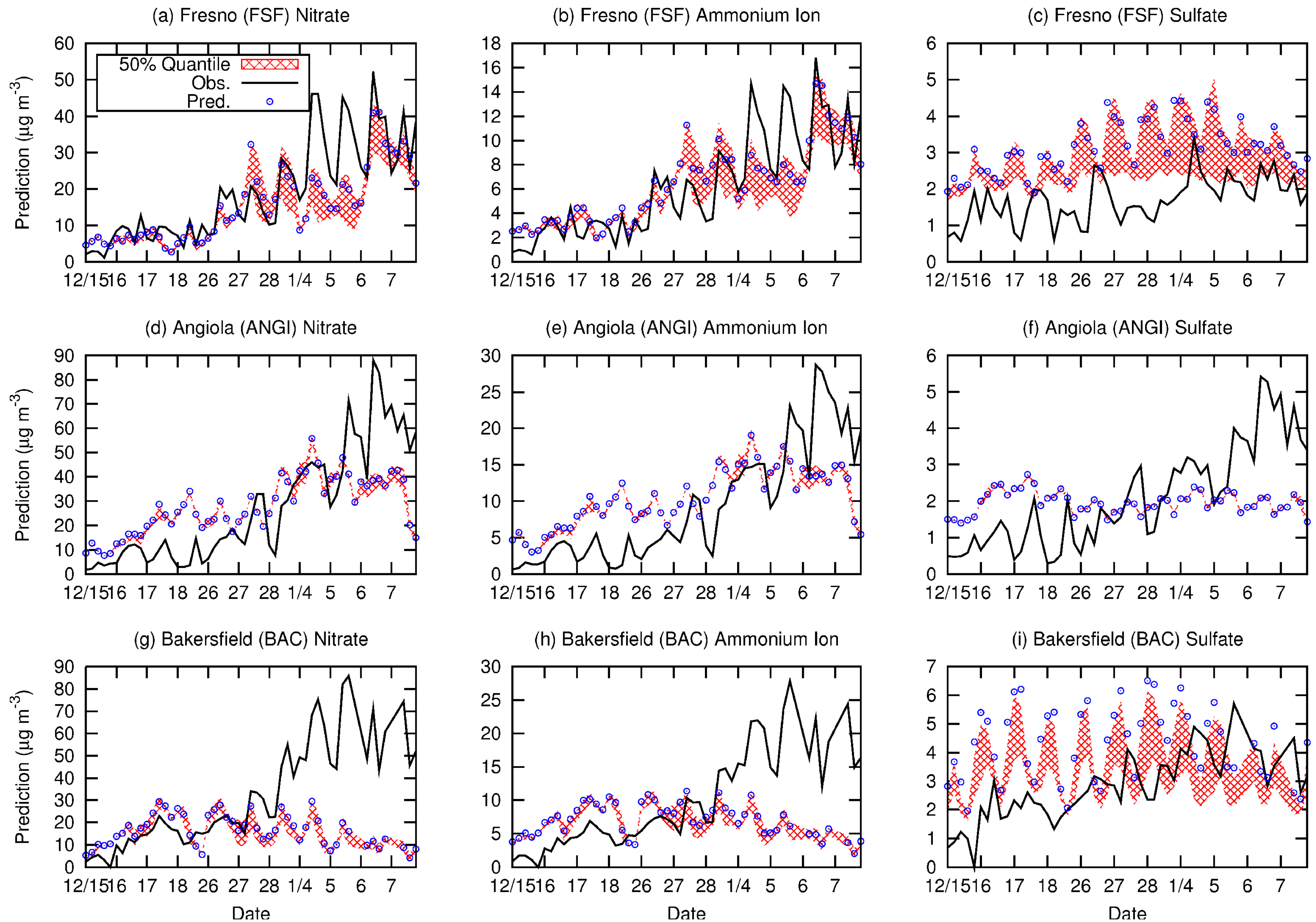
PM2.5 Nitrate, Ammonium, Sulfate



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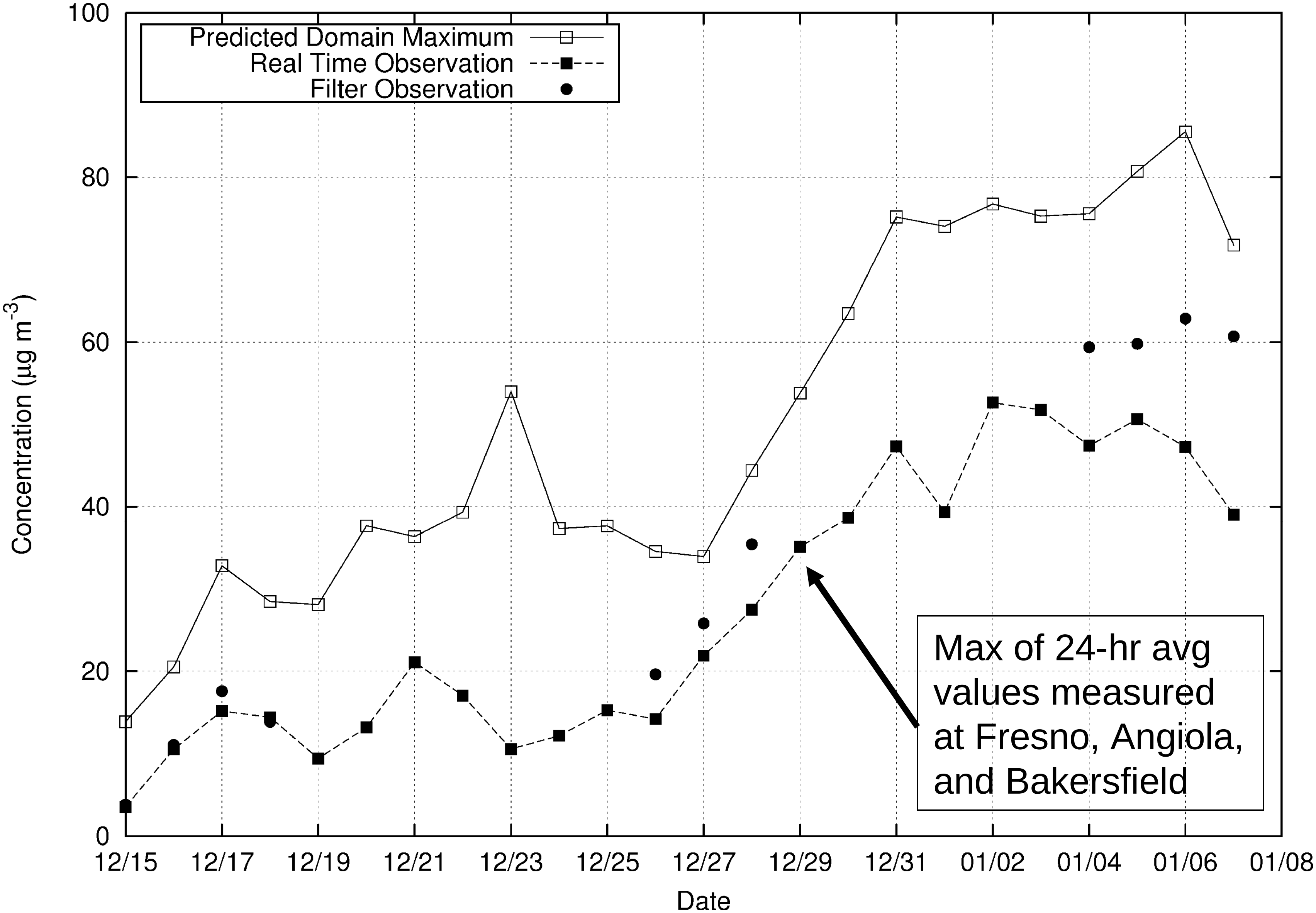
Predicted vs. Observed Nitrate Trends



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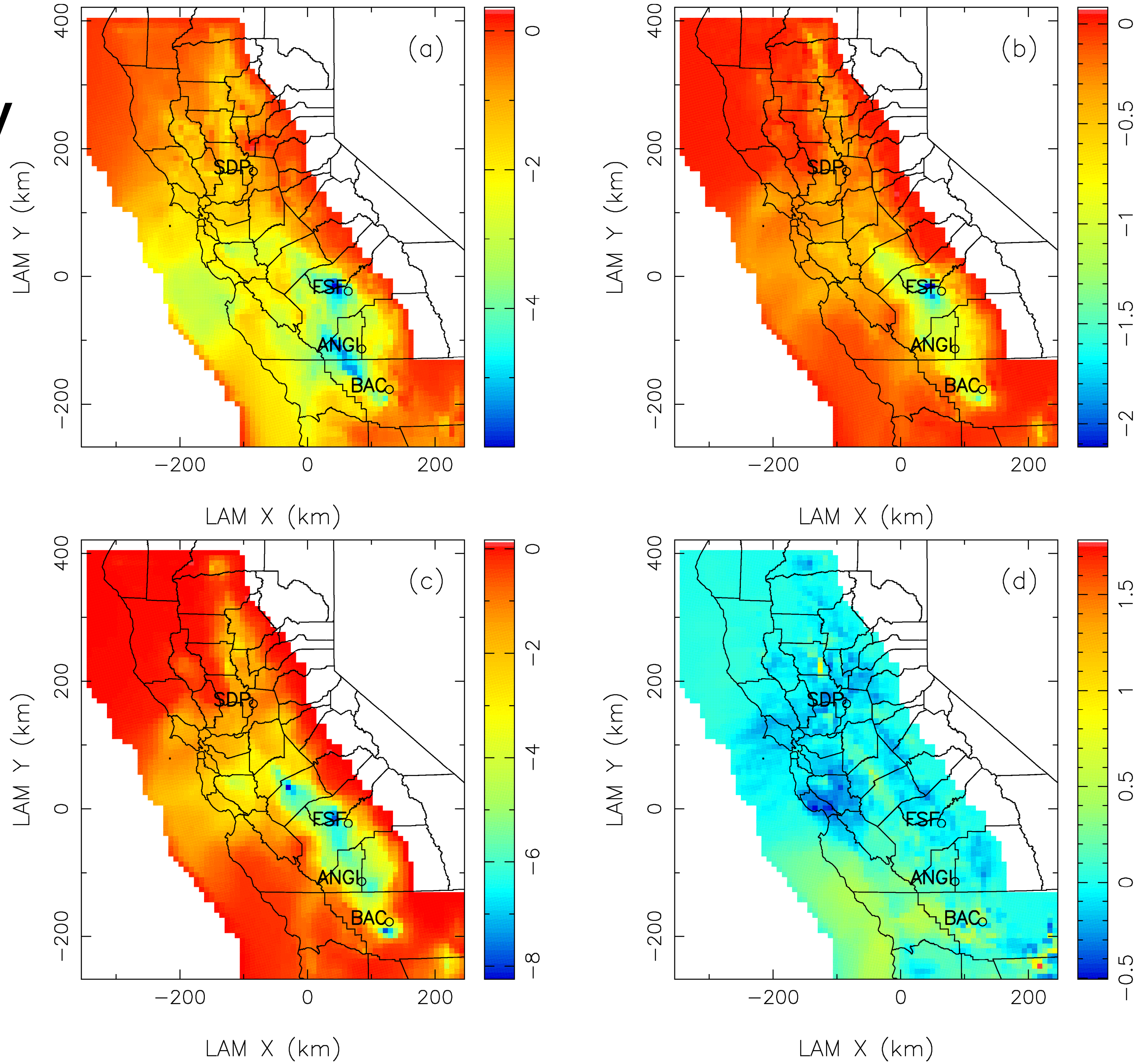
NO3 Sensitivity



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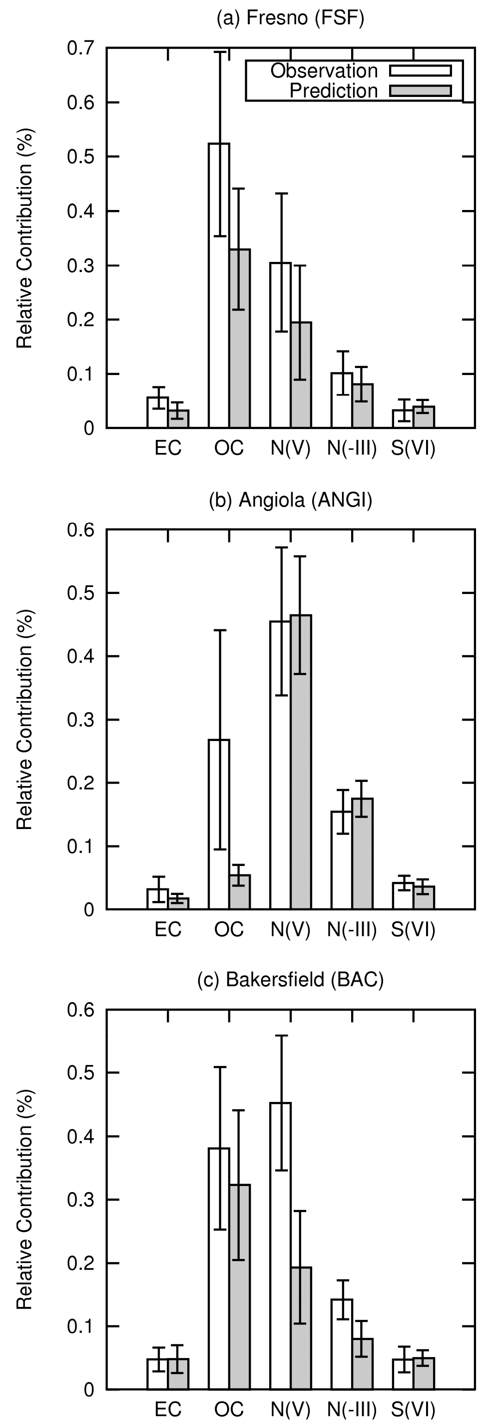
Relative Component Contributions to PM

Average and standard deviation of predictions and observations is based on 55 samples

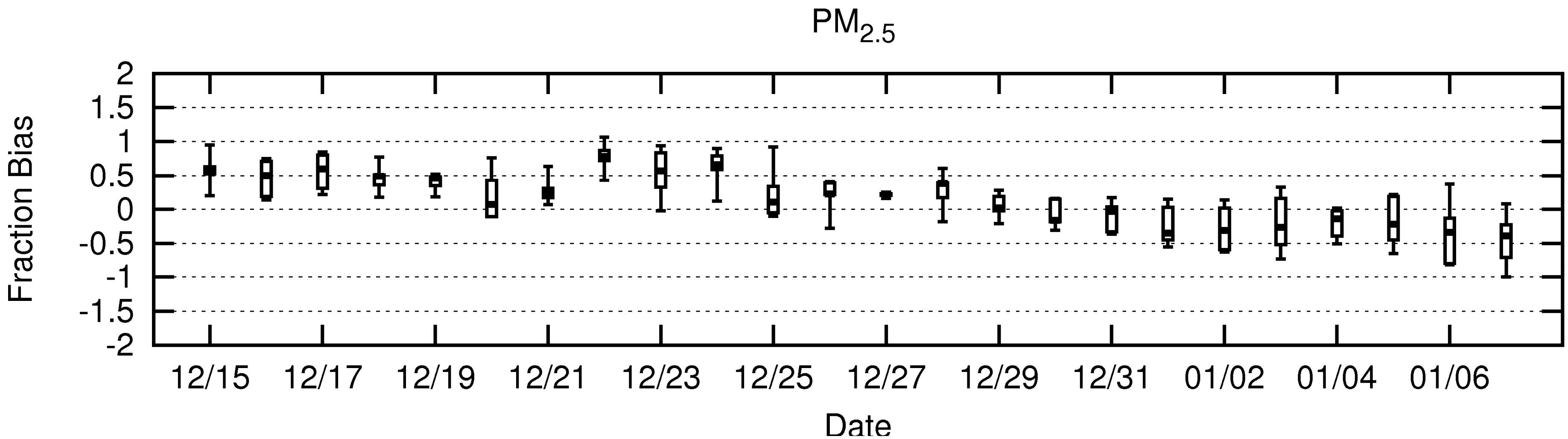
Urban locations (Fresno and Bakersfield)
Predictions and observations match except for nitrate under-prediction at Bakersfield (discussed previously)

Rural location (Angiola)
OC under-prediction. What primary sources are we missing? What SOA formation mechanisms are we missing?

Source: Q. Ying, J. Lu, P. Allen, P. Livingstone, A. Kaduwela, and M. Kleeman "Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part I. Base Case Model Results.", Atmos. Env., in press, 2008.



PM2.5 Fractional Bias



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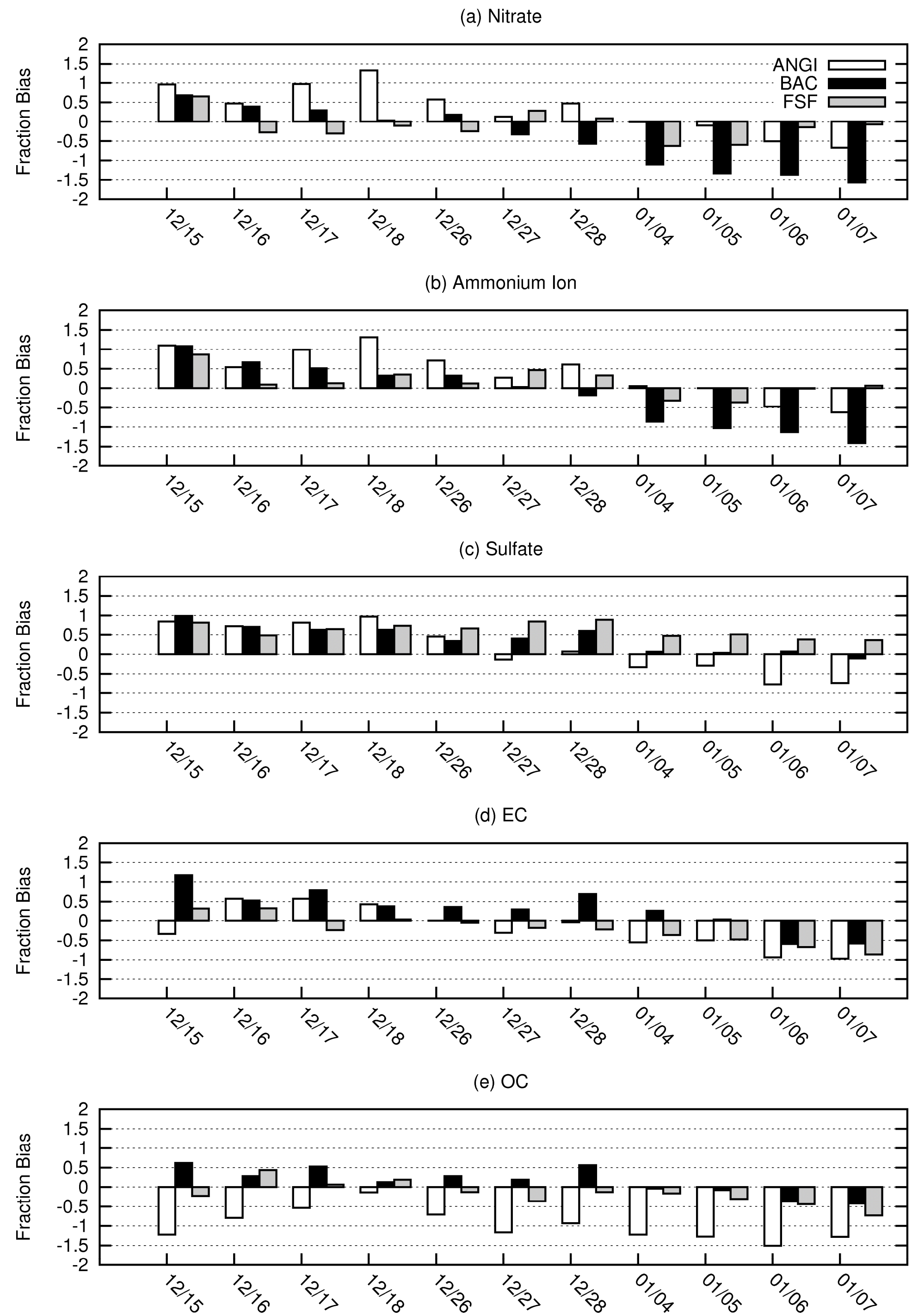
Fractional Bias of Individual PM2.5 Components

Most components – slight over-prediction during early portion of episode evolving to slight under-prediction later in episode

OC – always under-predicted at Angiola



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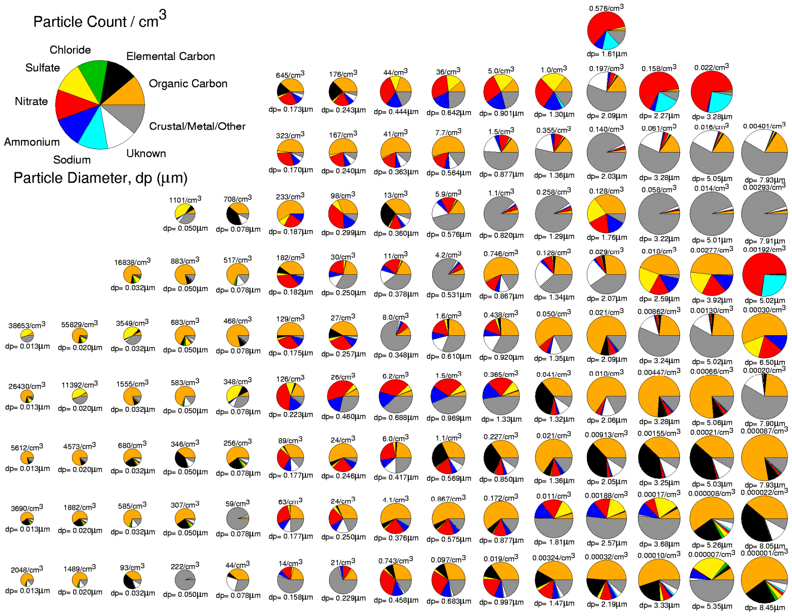
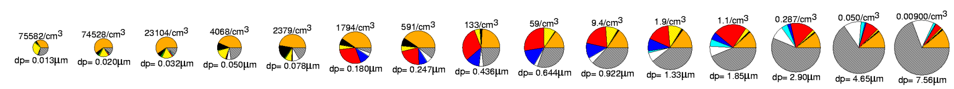
Source Apportionment Methodology

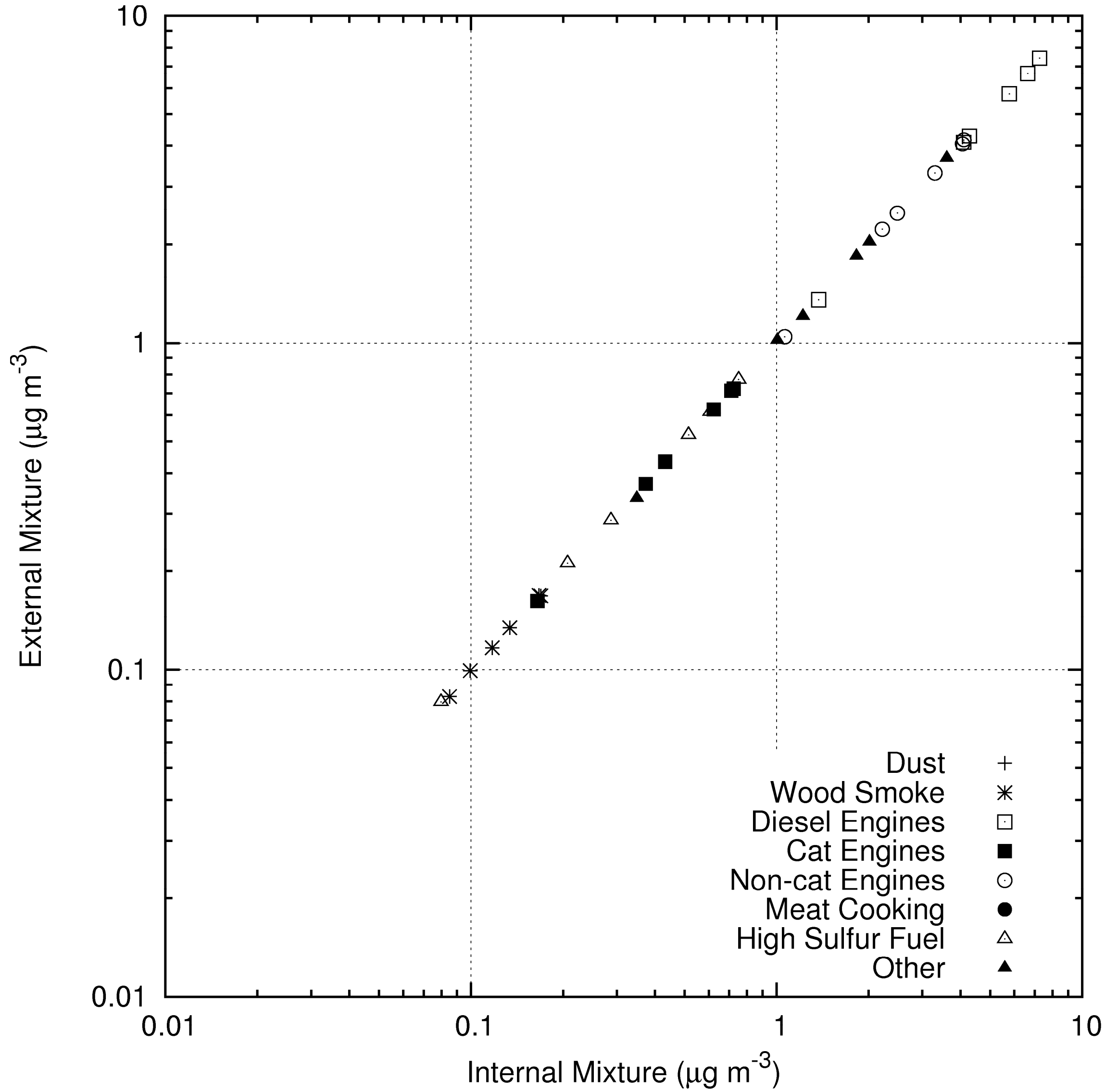
Source-oriented External Mixture



Internal Mixture With Artificial Tracers



Internal Vs. External Mixture Comparison



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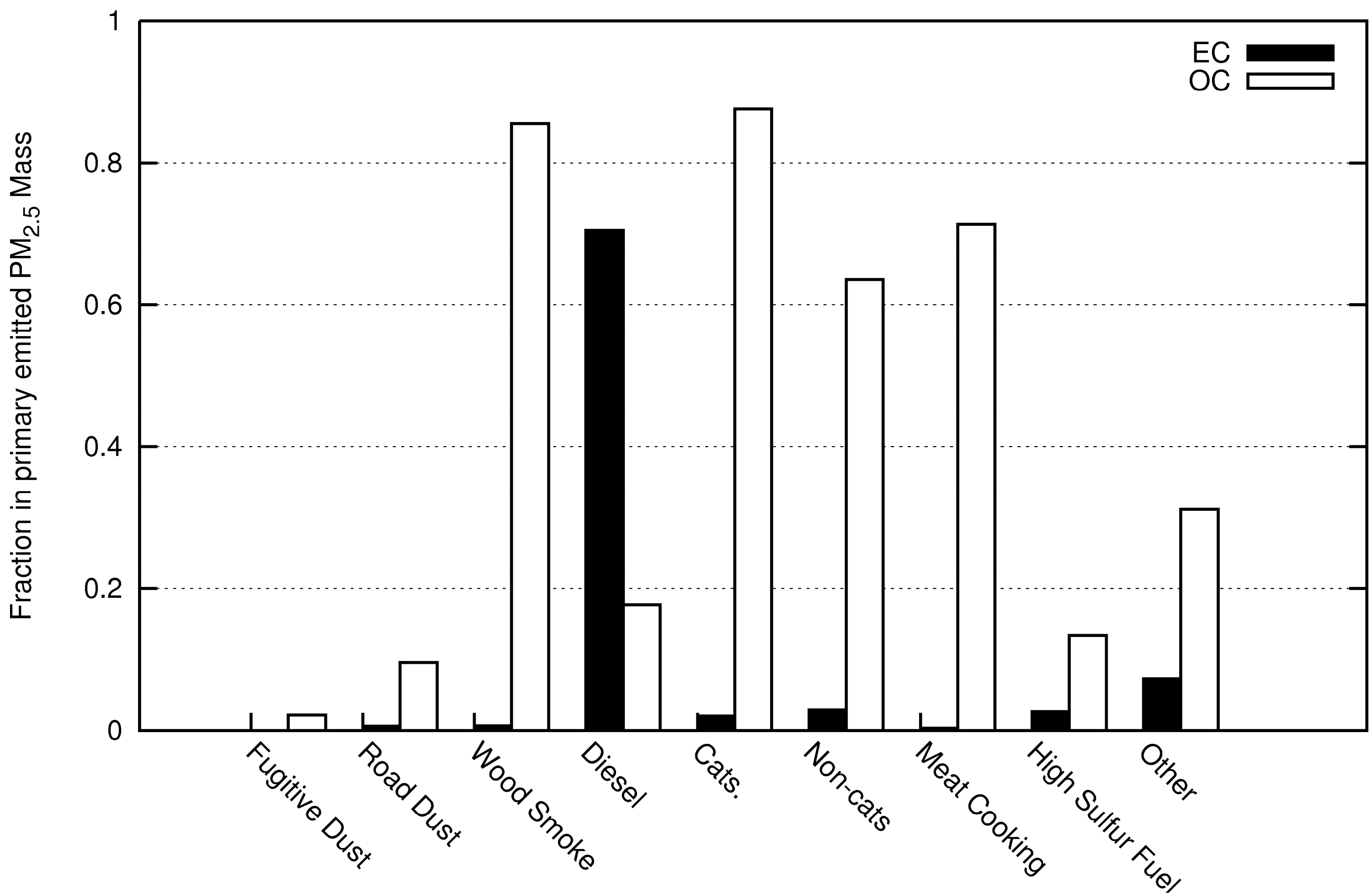
Averaged PM2.5 EC and OC Fractions For Internally Mixed Source Apportionment



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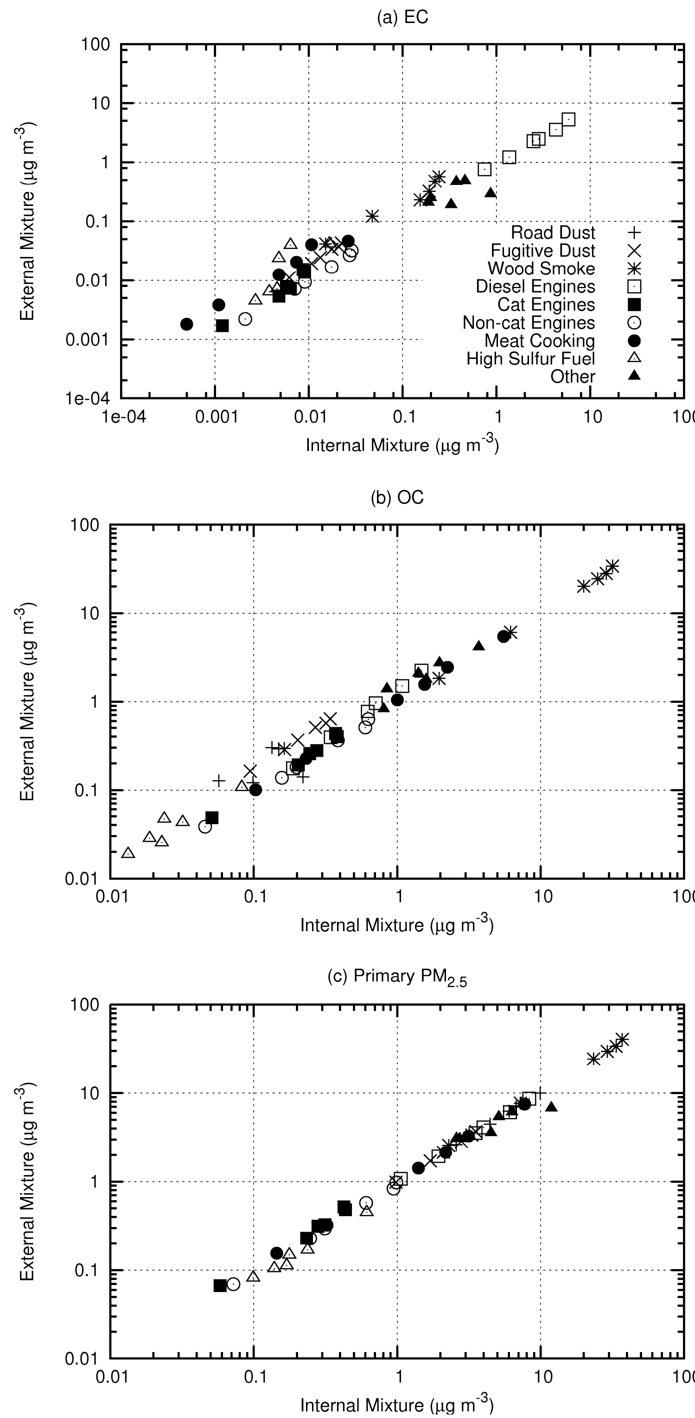
Internal vs. External Mixture Comparison

Relatively good agreement above 1 μg m^{-3}



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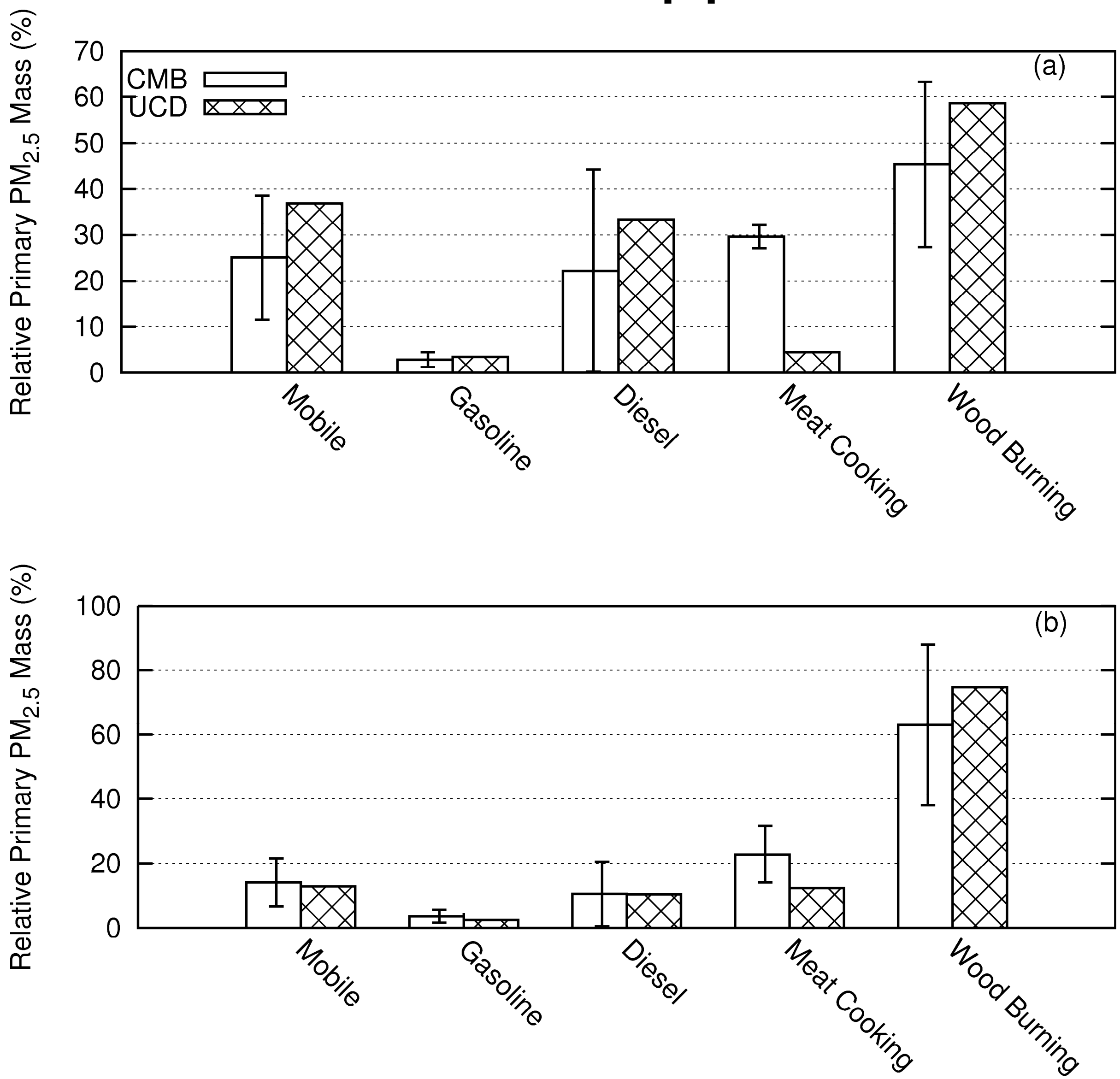
Grid Model vs. CMB Source Apportionment

Angiola

**Dust sources removed from grid model

Fresno

**Dust sources removed from grid model



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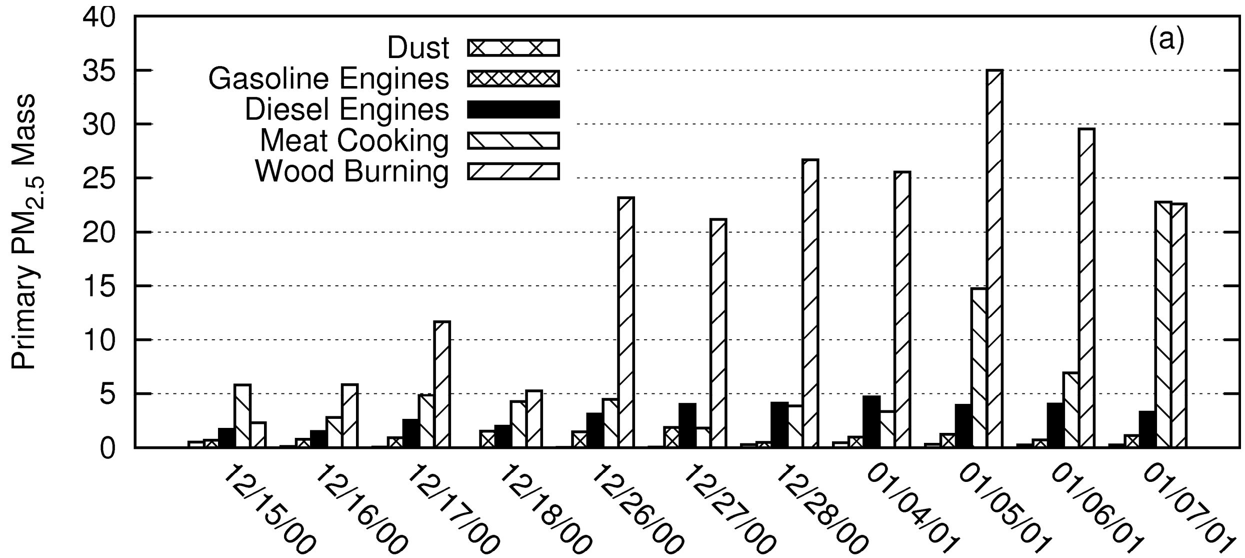
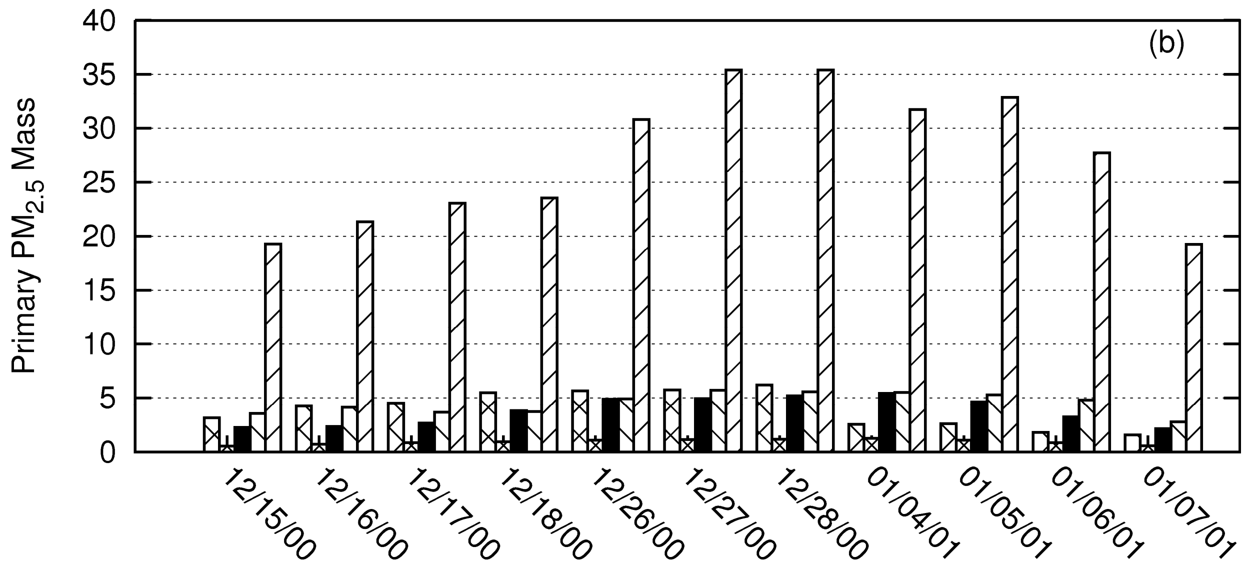
Fresno Grid Model vs. CMB Source Apportionment

CMB

**these results do not match longer averaging time shown on previous slide



Grid Model



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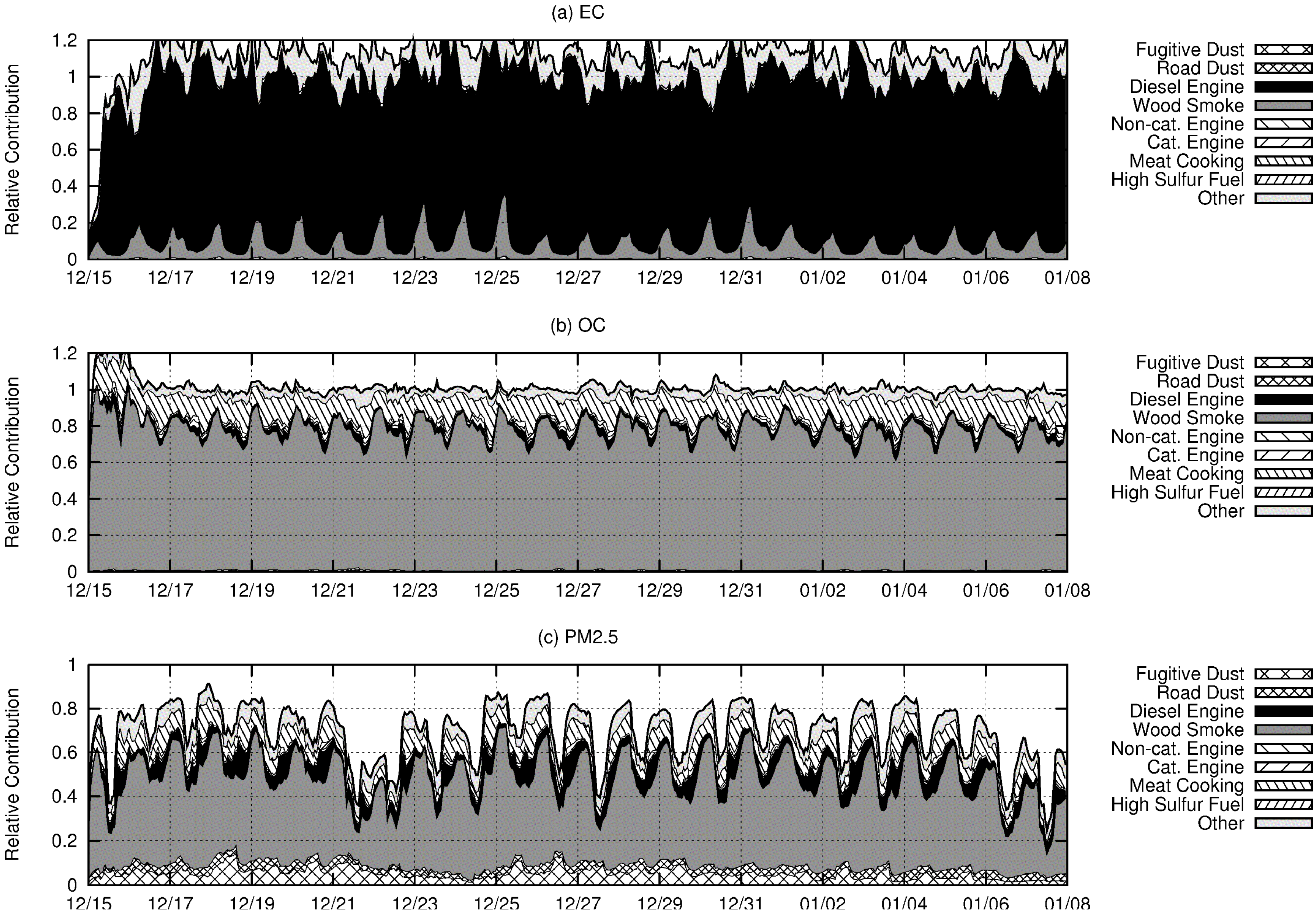
Fresno PM2.5 Source Contributions



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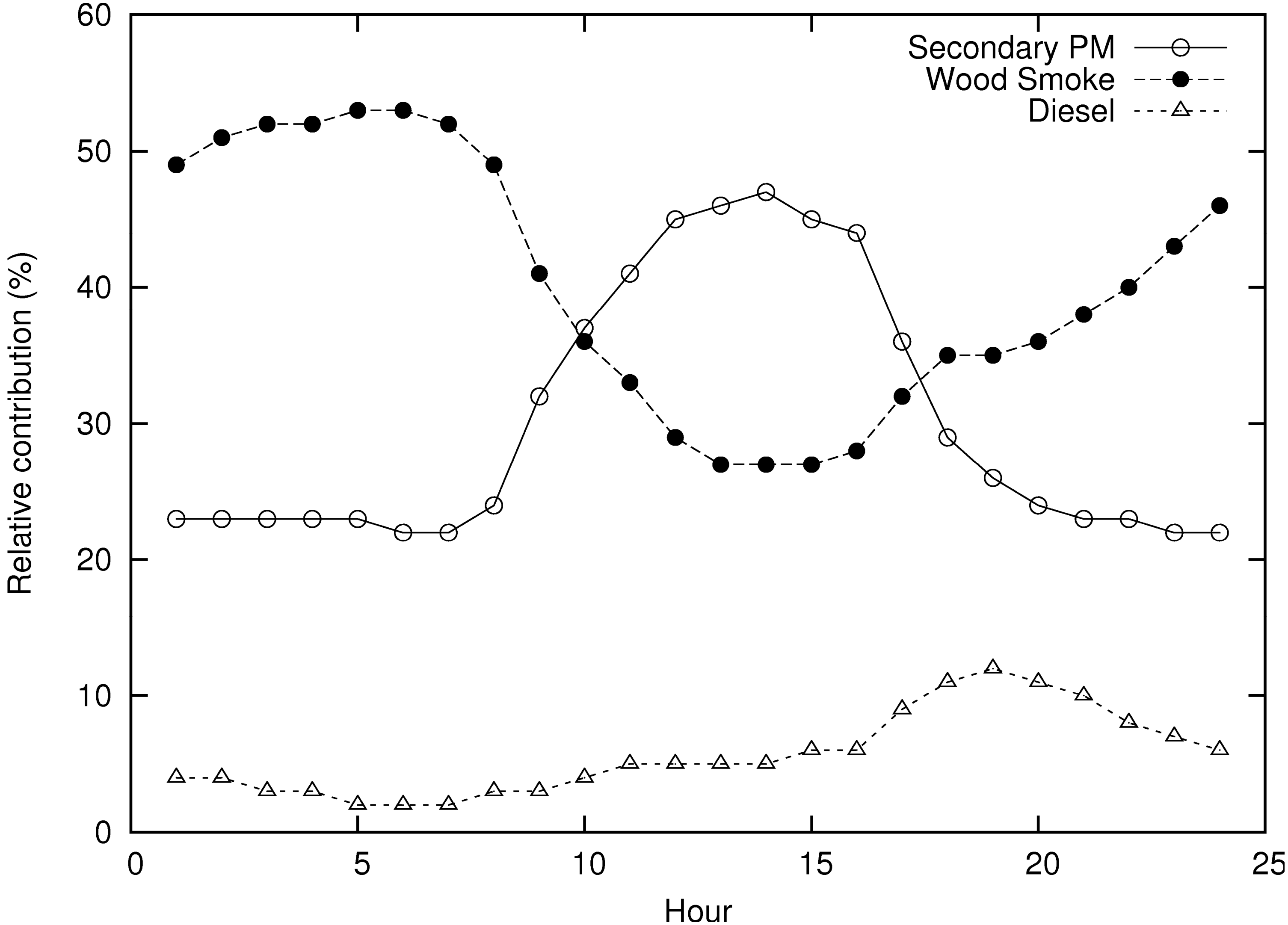
Average Diurnal Variation of Source Contributions



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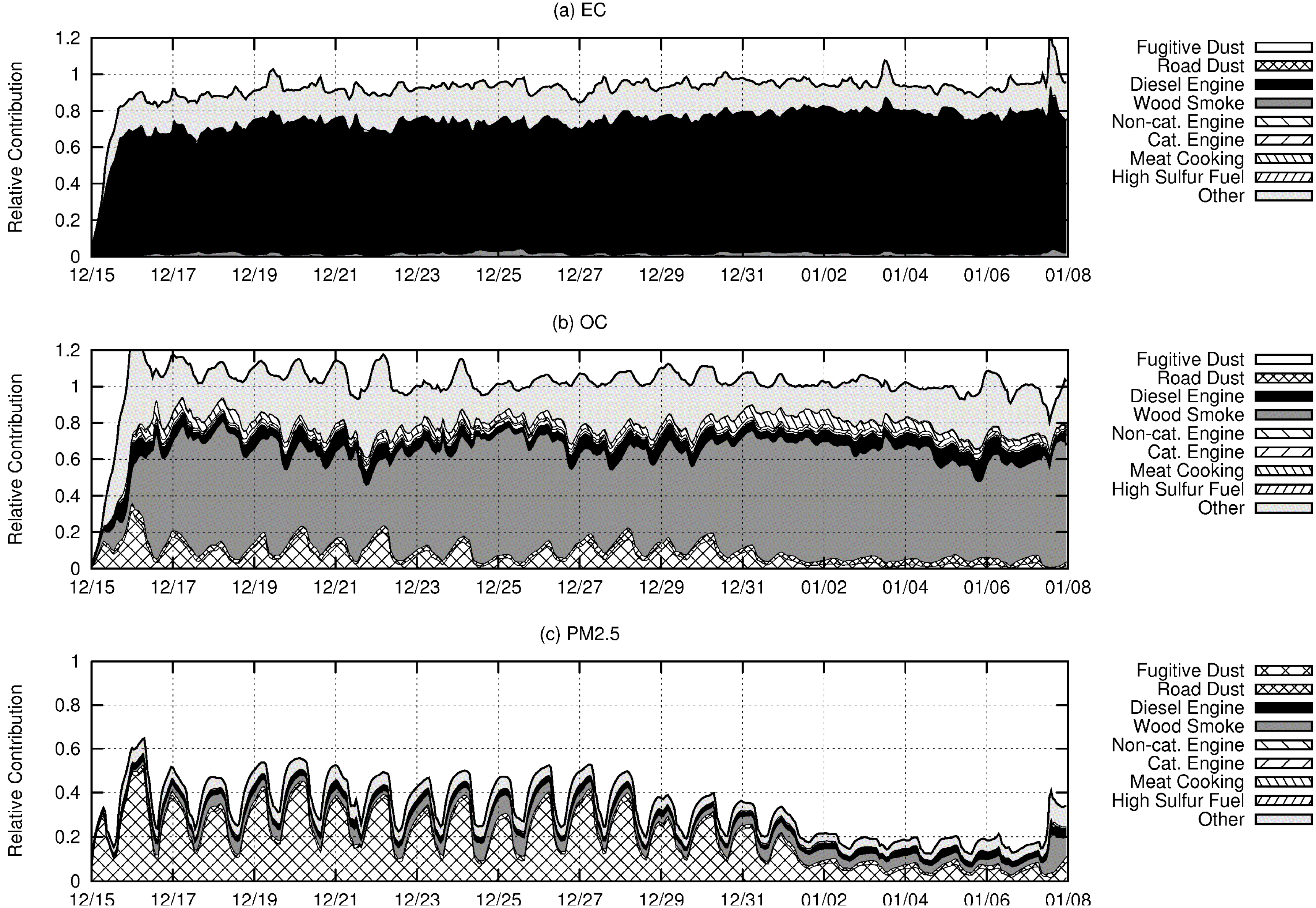
Angiola PM2.5 Source Contributions



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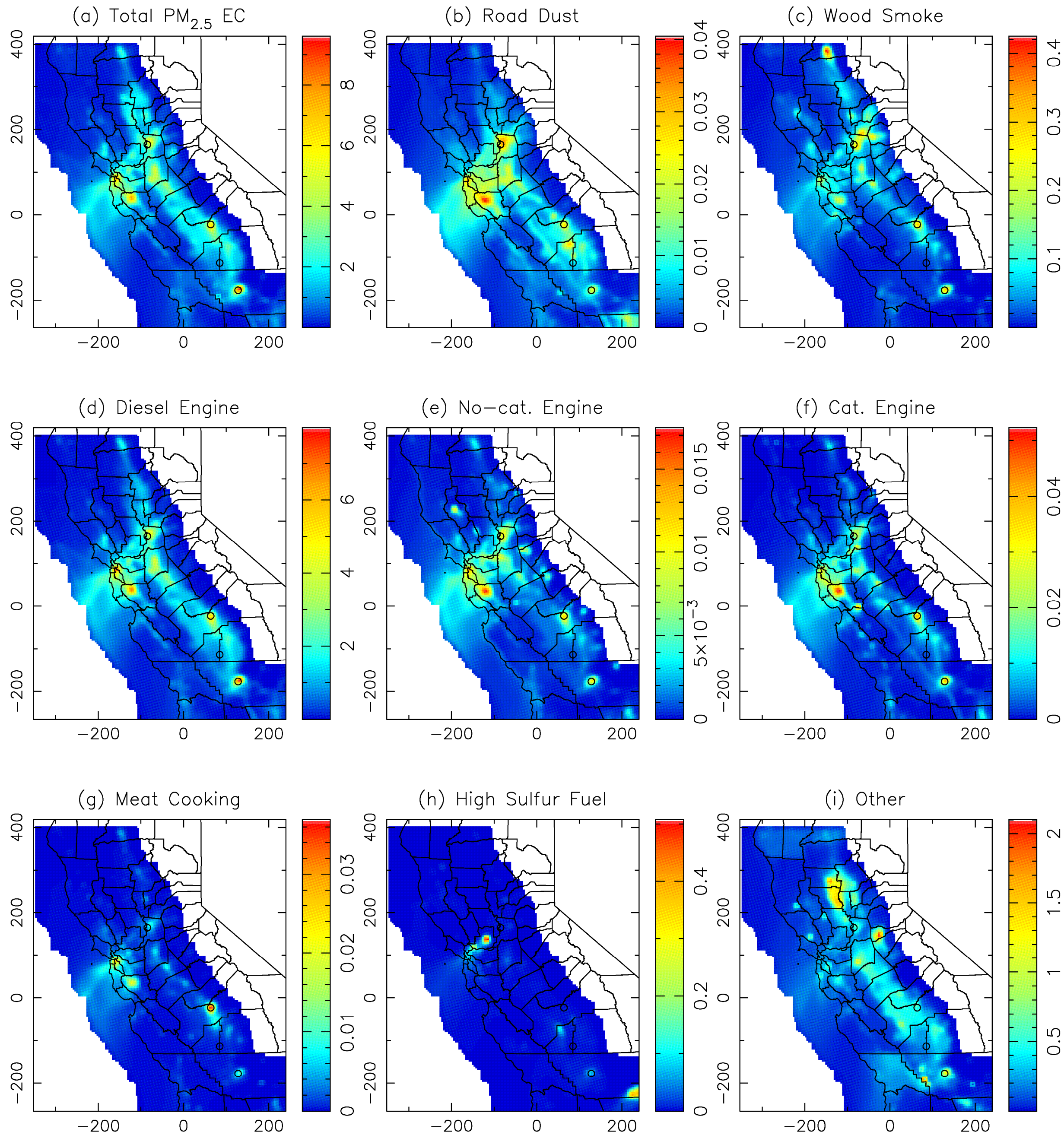
Regional EC Source Contributions



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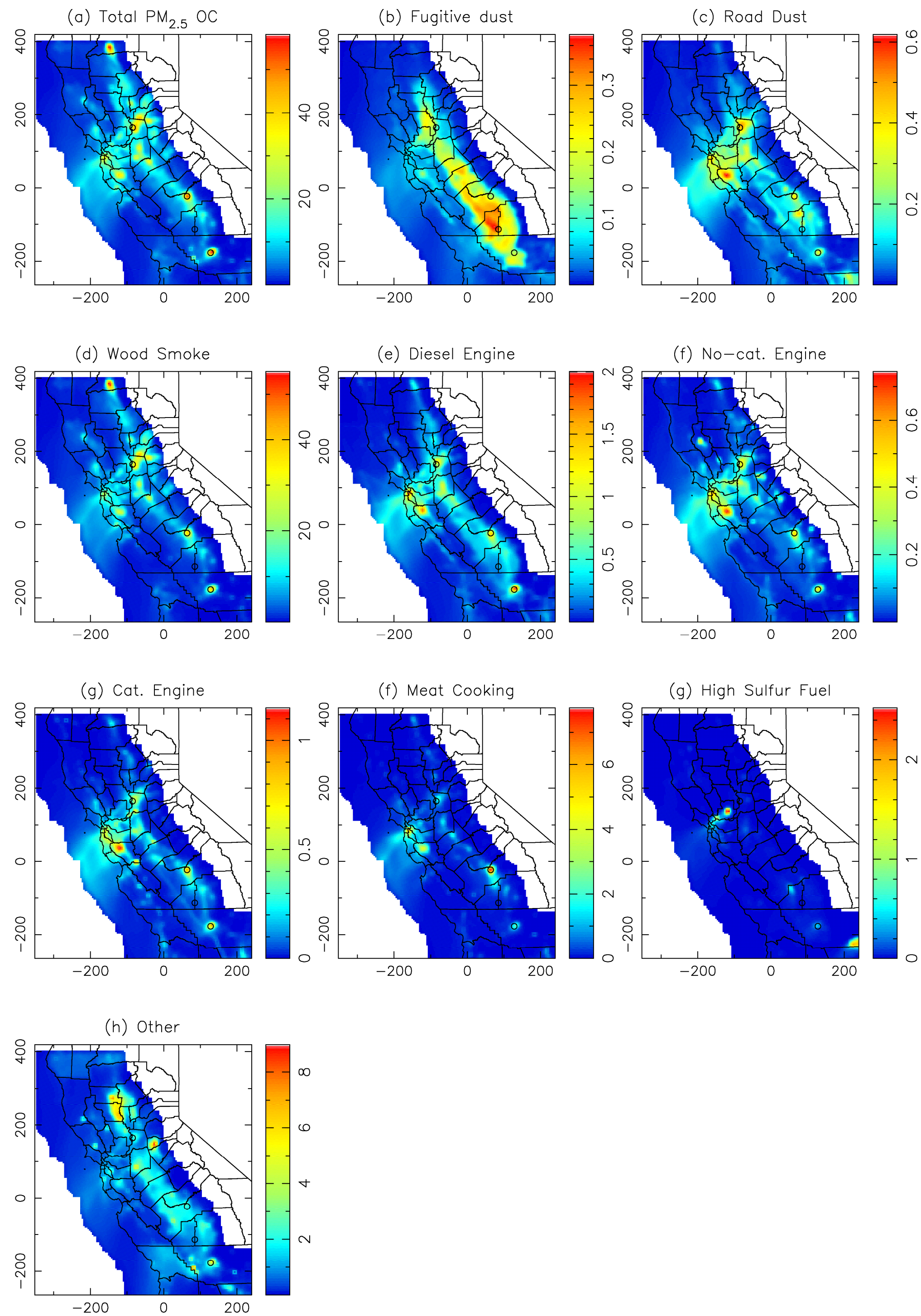
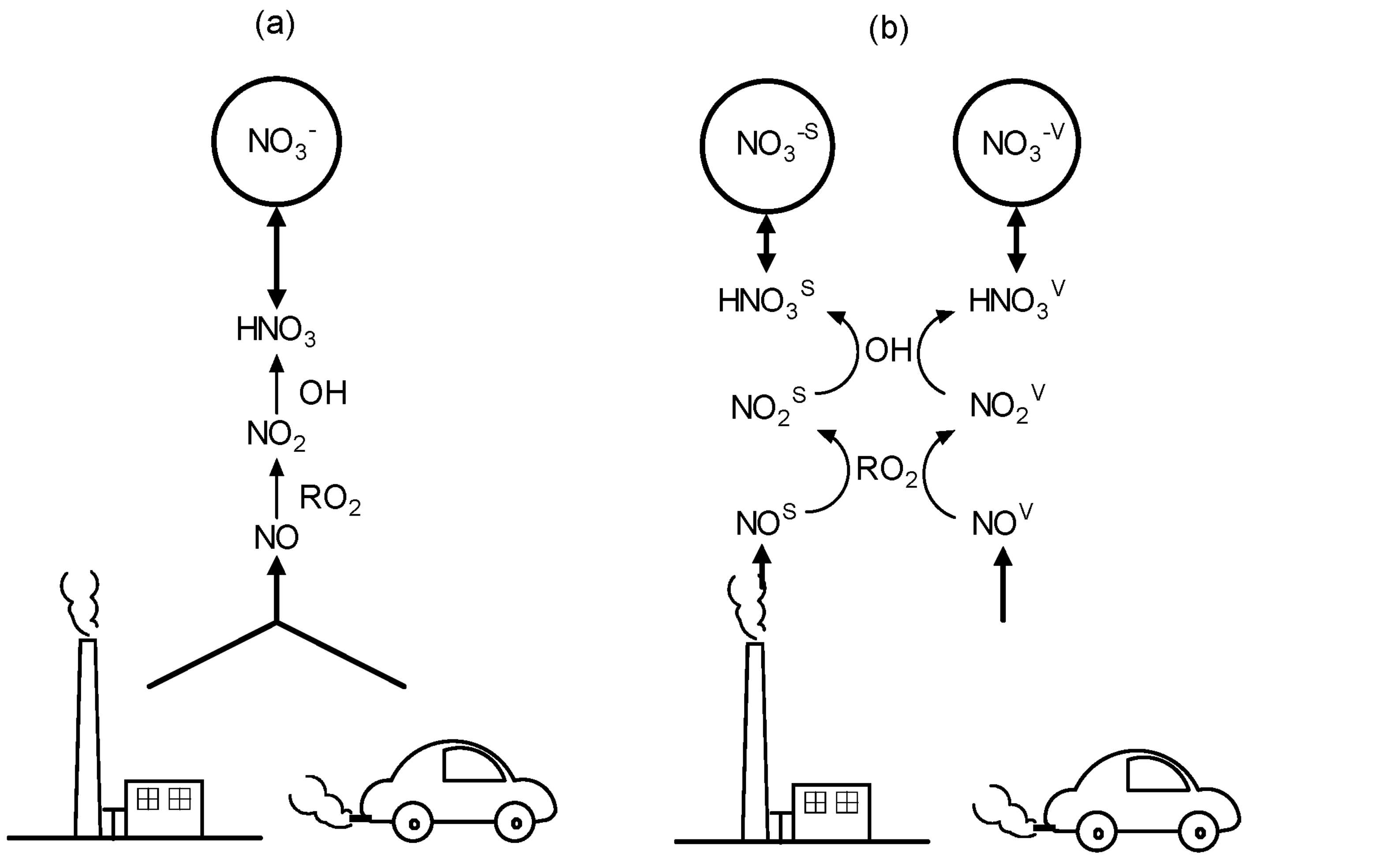
Regional OC Source Contributions



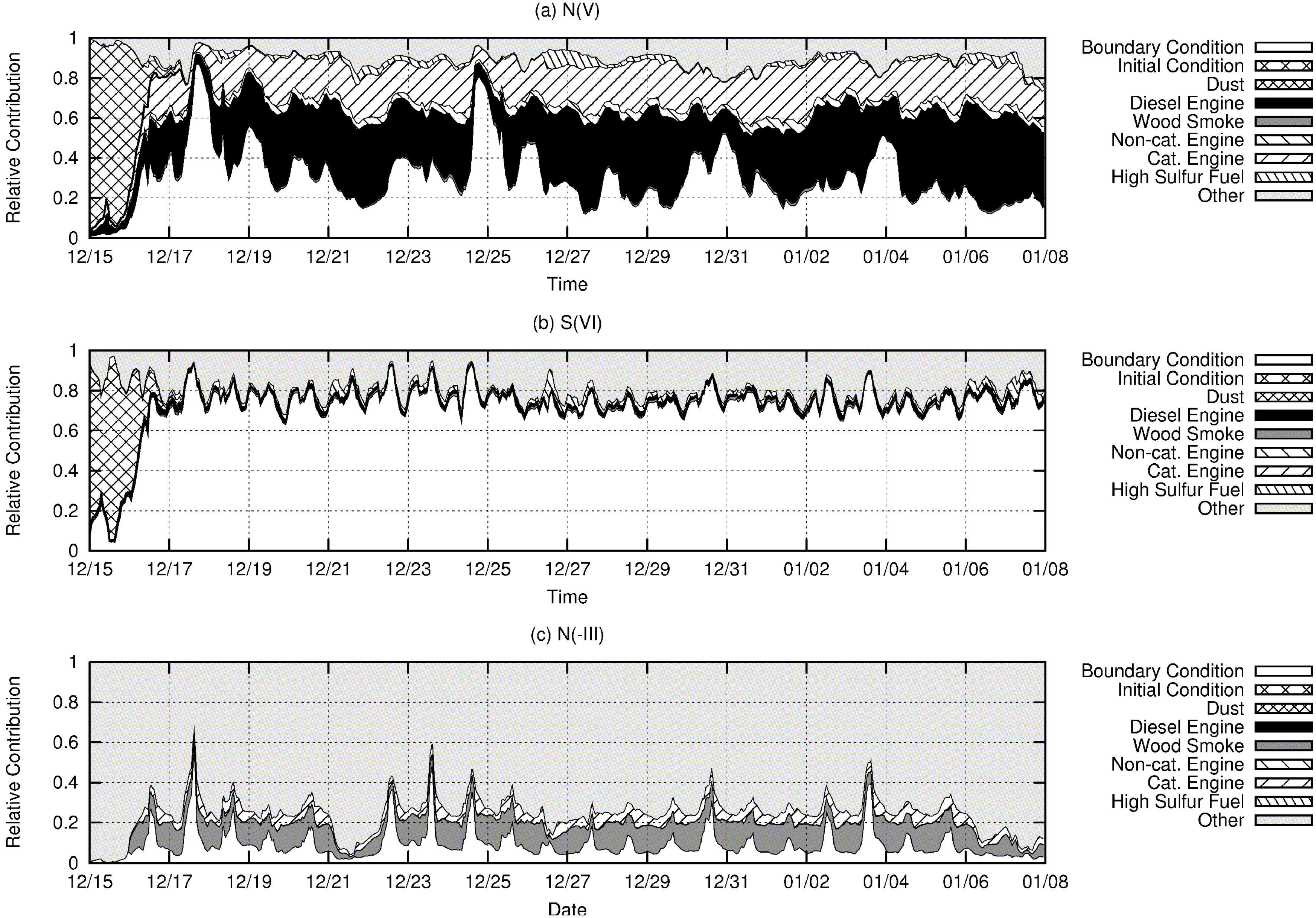
Source: Q. Ying, J. Lu, A. Kaduwela, and M. Kleeman "Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part II. Regional Source Apportionment of Primary Airborne Particulate Matter.", Atmos. Env., in press, 2008.

Source Apportionment of Secondary PM



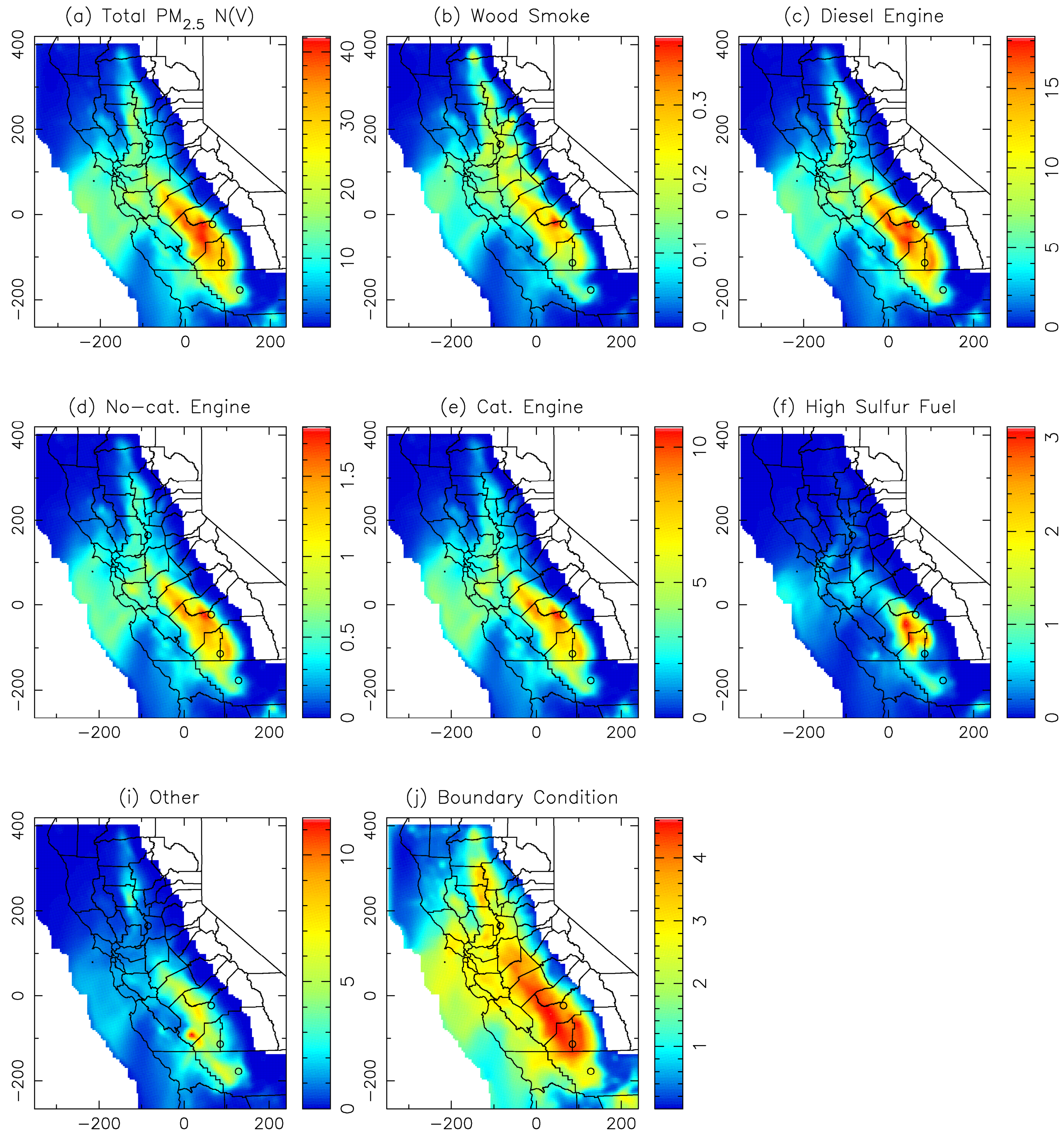
Source: Ying, Q. and M.J. Kleeman. "Source contributions to the regional distribution of secondary particulate matter in California." Atmospheric Environment, Vol 40, pp 736-752, 2006.

Fresno Source Contributions

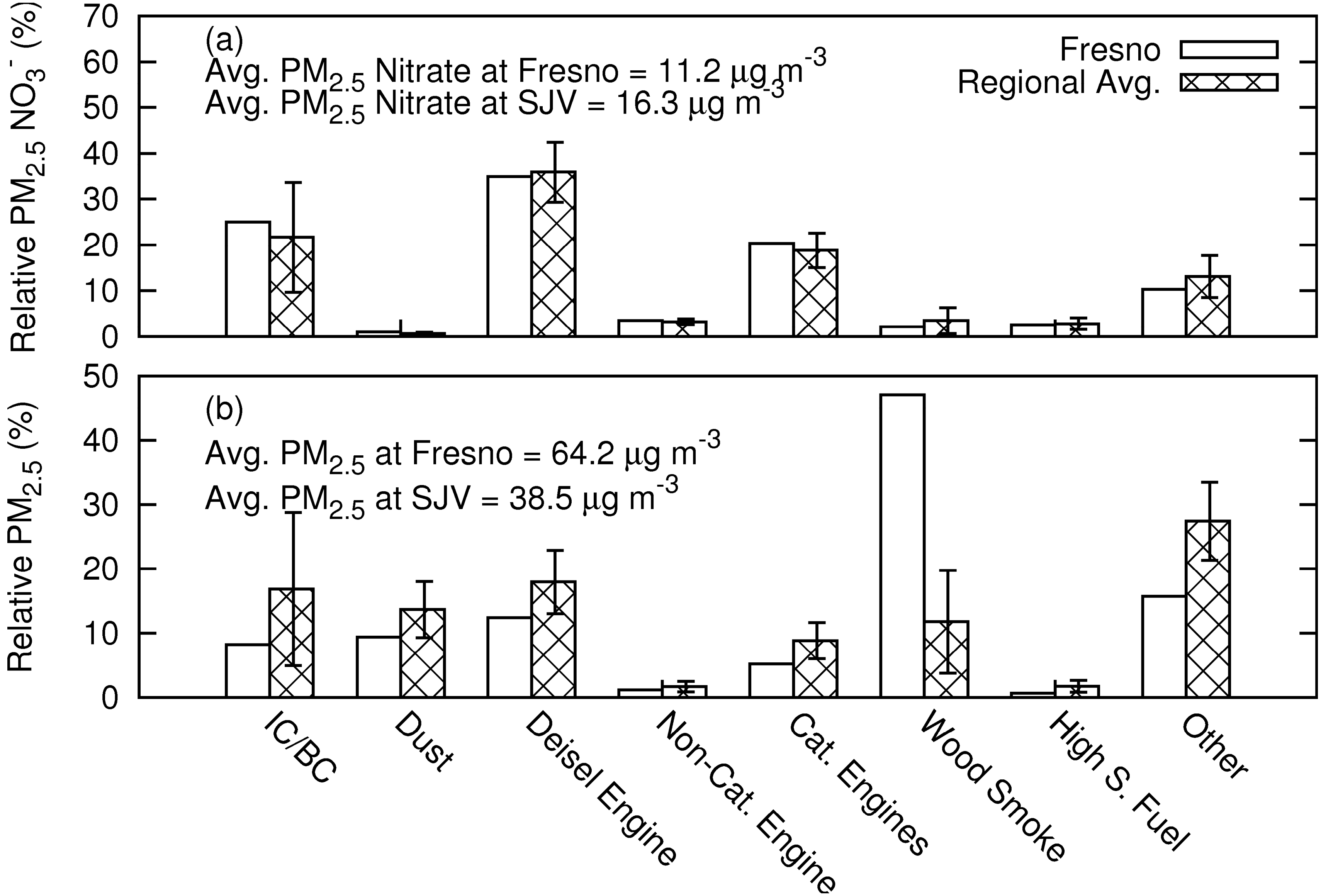


Source: Q. Ying, J. Lu, A. Kaduwela, and M. Kleeman "Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part III. Regional Source Apportionment of Secondary and Total Airborne PM2.5 and PM0.1.", Atmos. Env., in press, 2008.

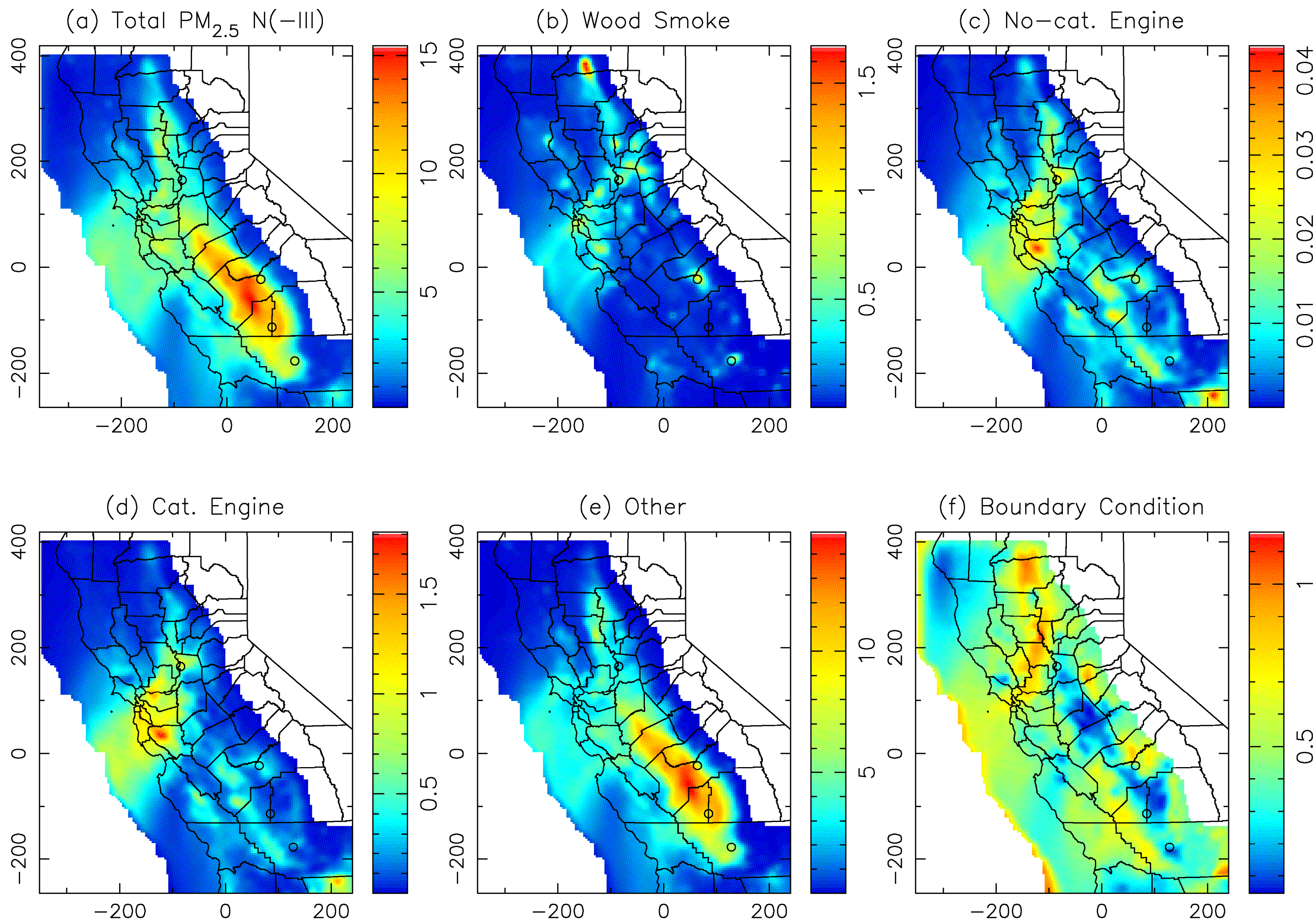
Regional Nitrate Source Contributions



Fresno vs. Region-wide Source Contributions

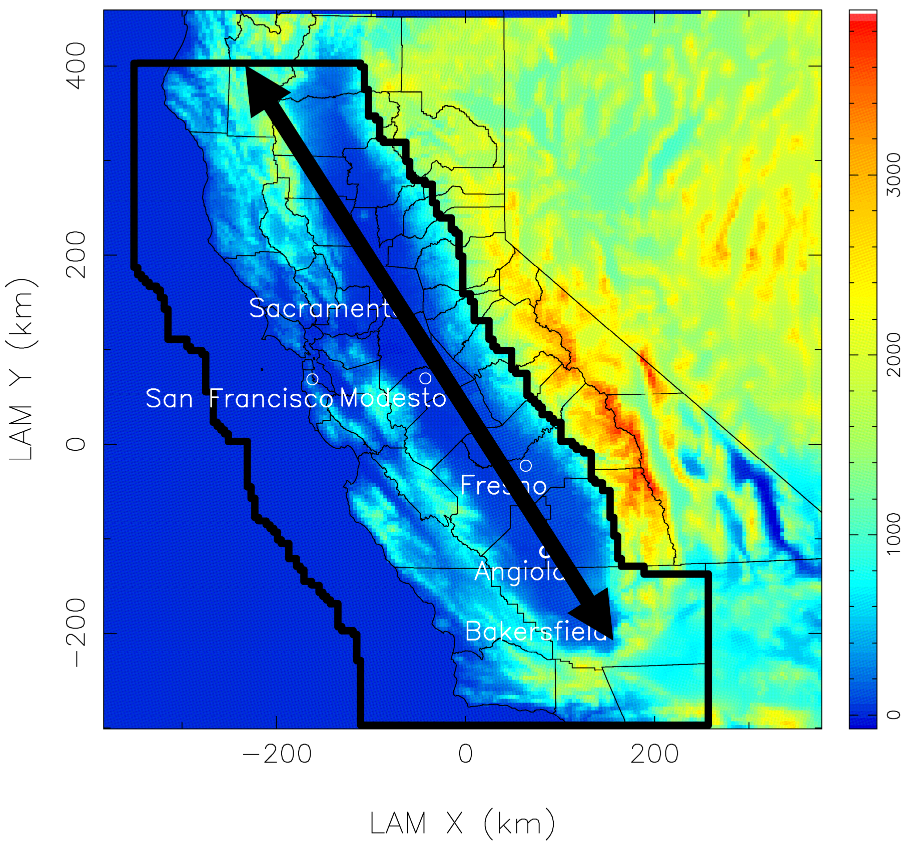


Regional NH4+ Source Contributions

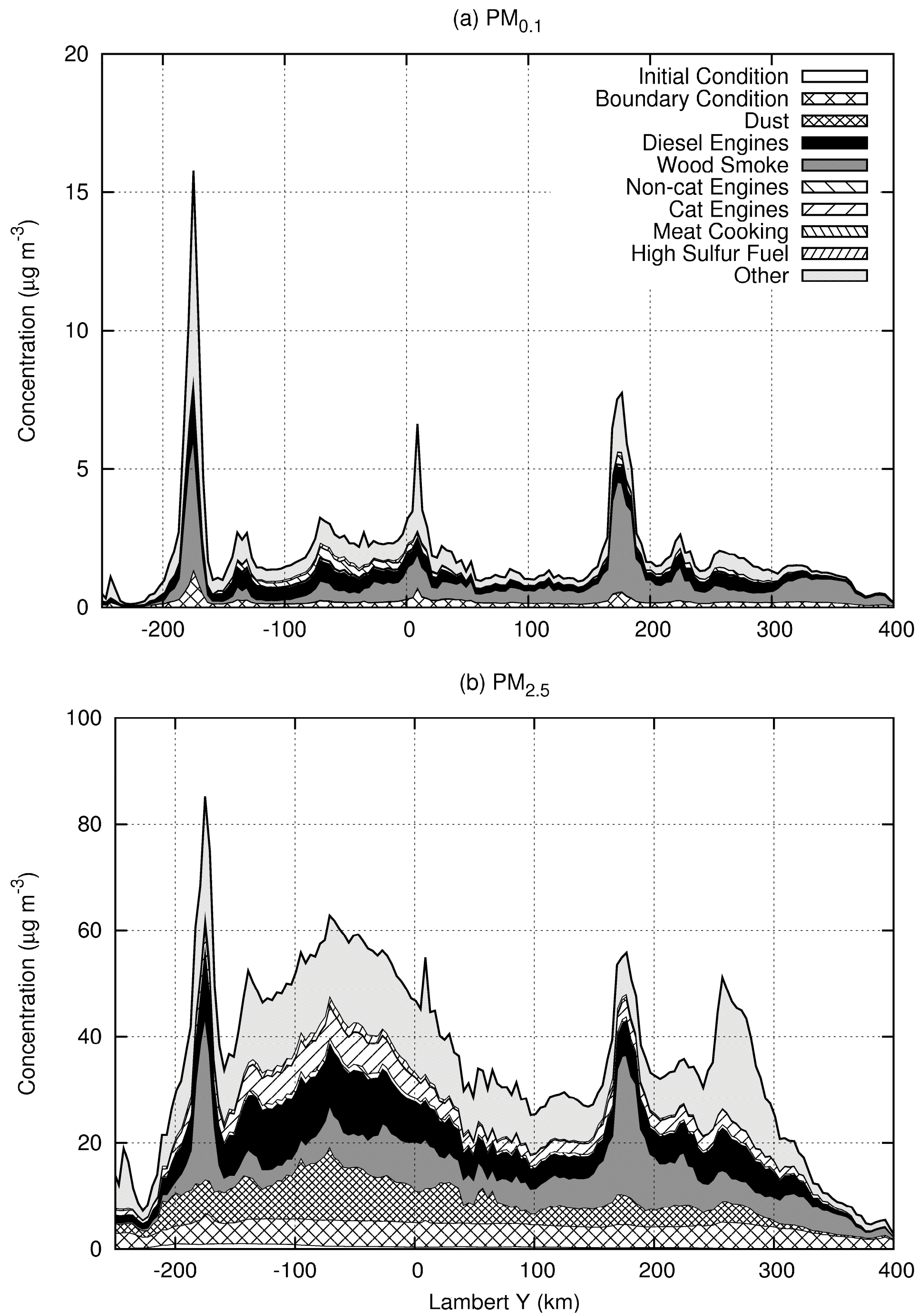


Source: Q. Ying, J. Lu, A. Kaduwela, and M. Kleeman “Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part III. Regional Source Apportionment of Secondary and Total Airborne PM2.5 and PM0.1.”, Atmos. Env., in press, 2008.

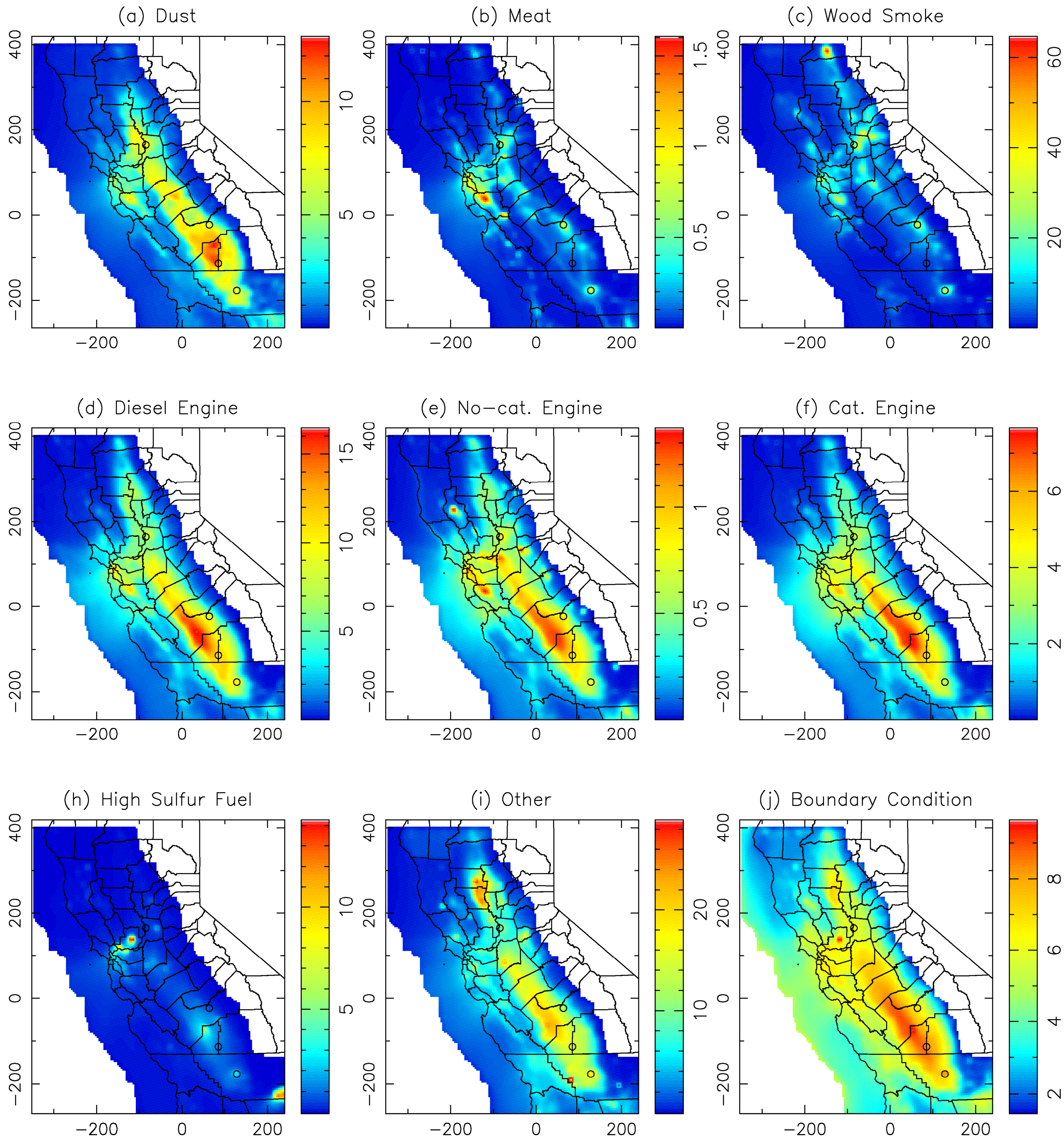
PM Spatial Gradients



Source: Q. Ying, J. Lu, A. Kaduwela, and M. Kleeman "Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part III. Regional Source Apportionment of Secondary and Total Airborne PM2.5 and PM0.1.", Atmos. Env., in press, 2008.



Regional PM2.5 (primary + secondary) Source Contributions



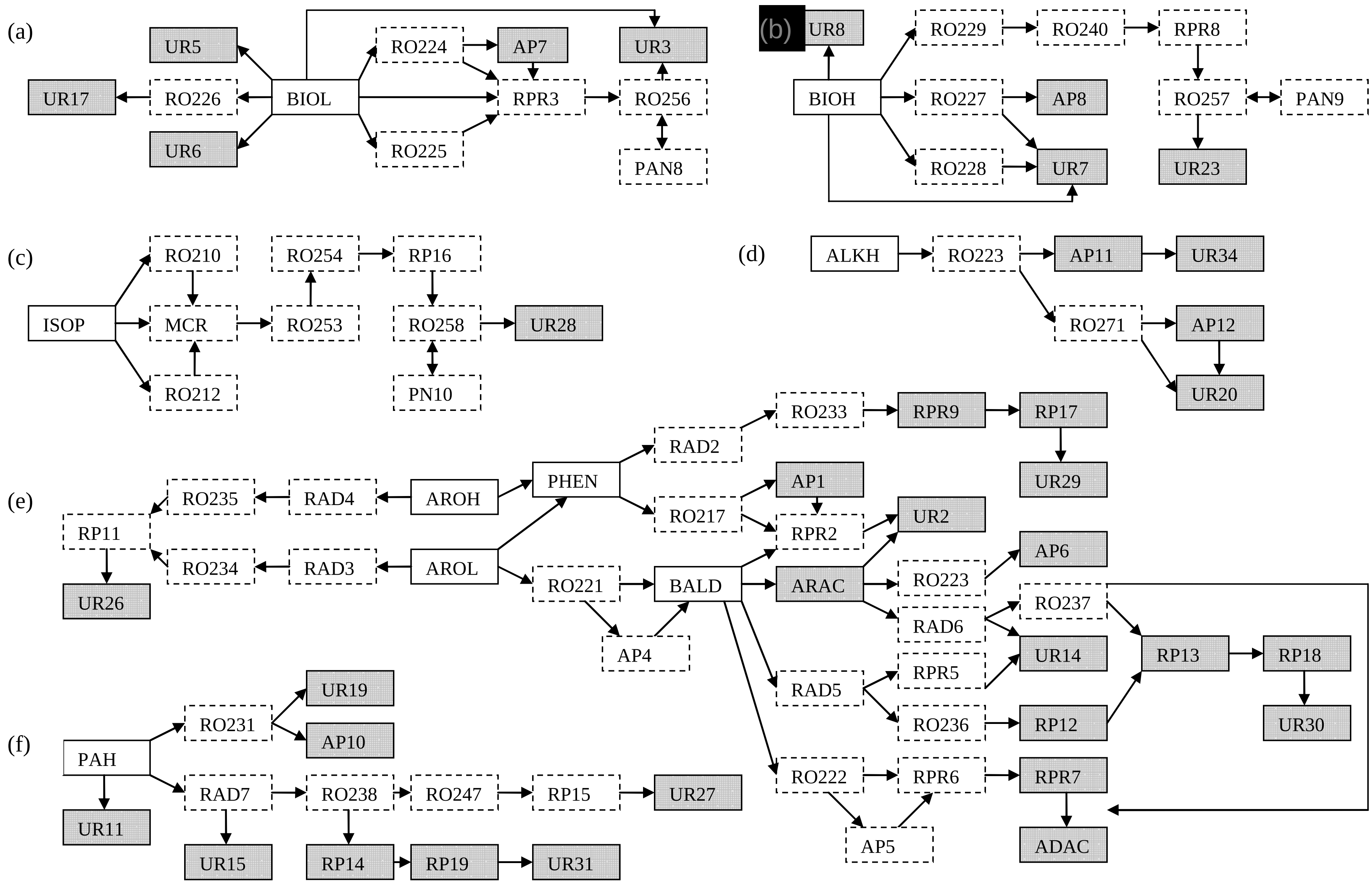
Source: Q. Ying, J. Lu, A. Kaduwela, and M. Kleeman "Modeling Air Quality During the California Regional PM10/PM2.5 Air Quality Study (CRPAQS) Using the UCD/CIT Source-Oriented Air Quality Model – Part III. Regional Source Apportionment of Secondary and Total Airborne PM2.5 and PM0.1.", Atmos. Env., in press, 2008.

SOA Source Apportionment

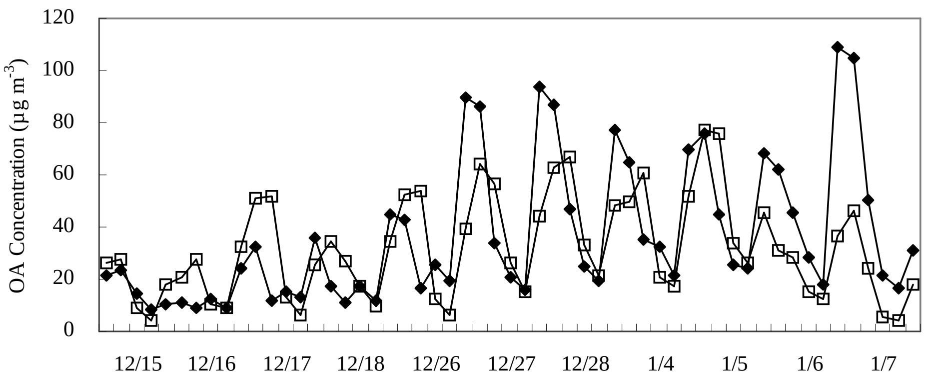
- Same techniques used for nitrate source apportionment can also be applied to SOA

Caltech Atmospheric Chemistry Mechanism (CACM)

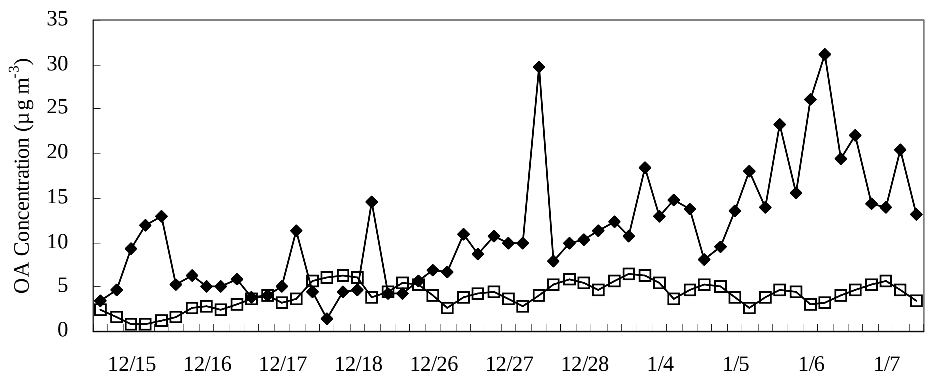
- Designed to simulate O3 and SOA formation
  - 210 species
  - 421 reactions
- Modifications for source apportionment of SOA
  - 1027 species
  - 2038 reactions


(a)
UR5
UR17
RO226
BIOL
UR6
RO224
AP7
RPR3
RO225
UR3
RO256
PAN8
(b)
UR8
BIOH
RO229
RO240
RPR8
RO227
AP8
RO257
PAN9
RO228
UR7
UR23
(c)
ISOP
RO210
MCR
RO212
RO254
RO253
RP16
RO258
UR28
PN10
(d)
ALKH
RO223
AP11
UR34
RO271
AP12
UR20
(e)
RP11
RO235
RO234
UR26
RAD4
RAD3
AROH
AROL
PHEN
RAD2
RO233
RPR9
RP17
UR29
RO217
AP1
RPR2
UR2
RO221
AP4
BALD
ARAC
RO223
AP6
RO237
RAD6
UR14
RP13
RP18
UR30
RAD5
RPR5
RO236
RP12
RO222
RPR6
RPR7
AP5
ADAC
(f)
PAH
UR11
RO231
UR19
AP10
RAD7
UR15
RO238
RO247
RP15
UR27
RP14
RP19
UR31

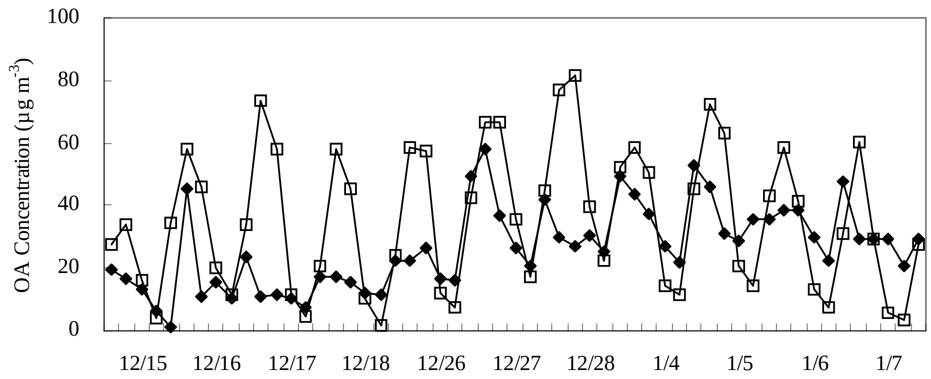
Predicted (open squares) and Observed OC (solid diamonds) using CACM



a.) Fresno

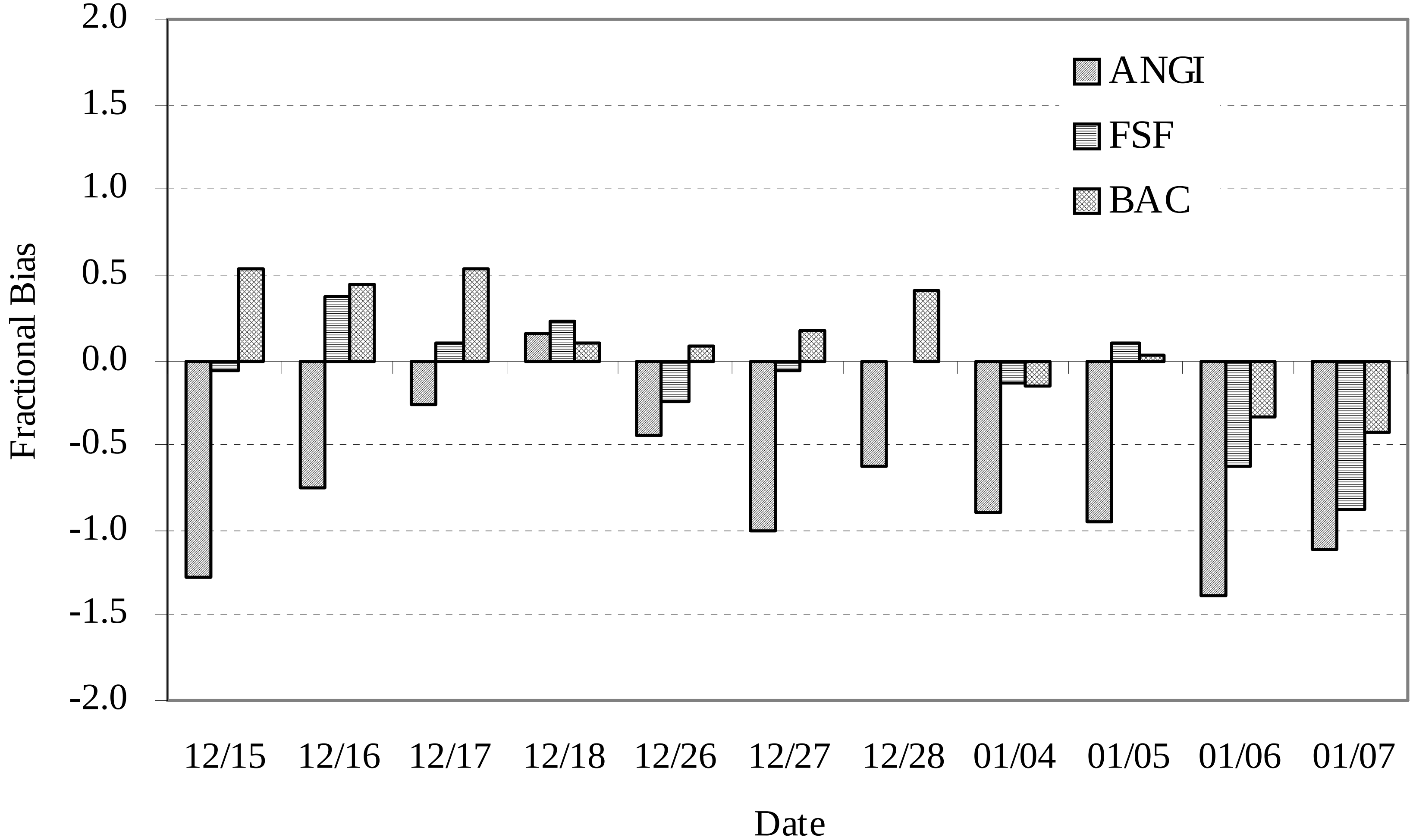


b.) Angiola

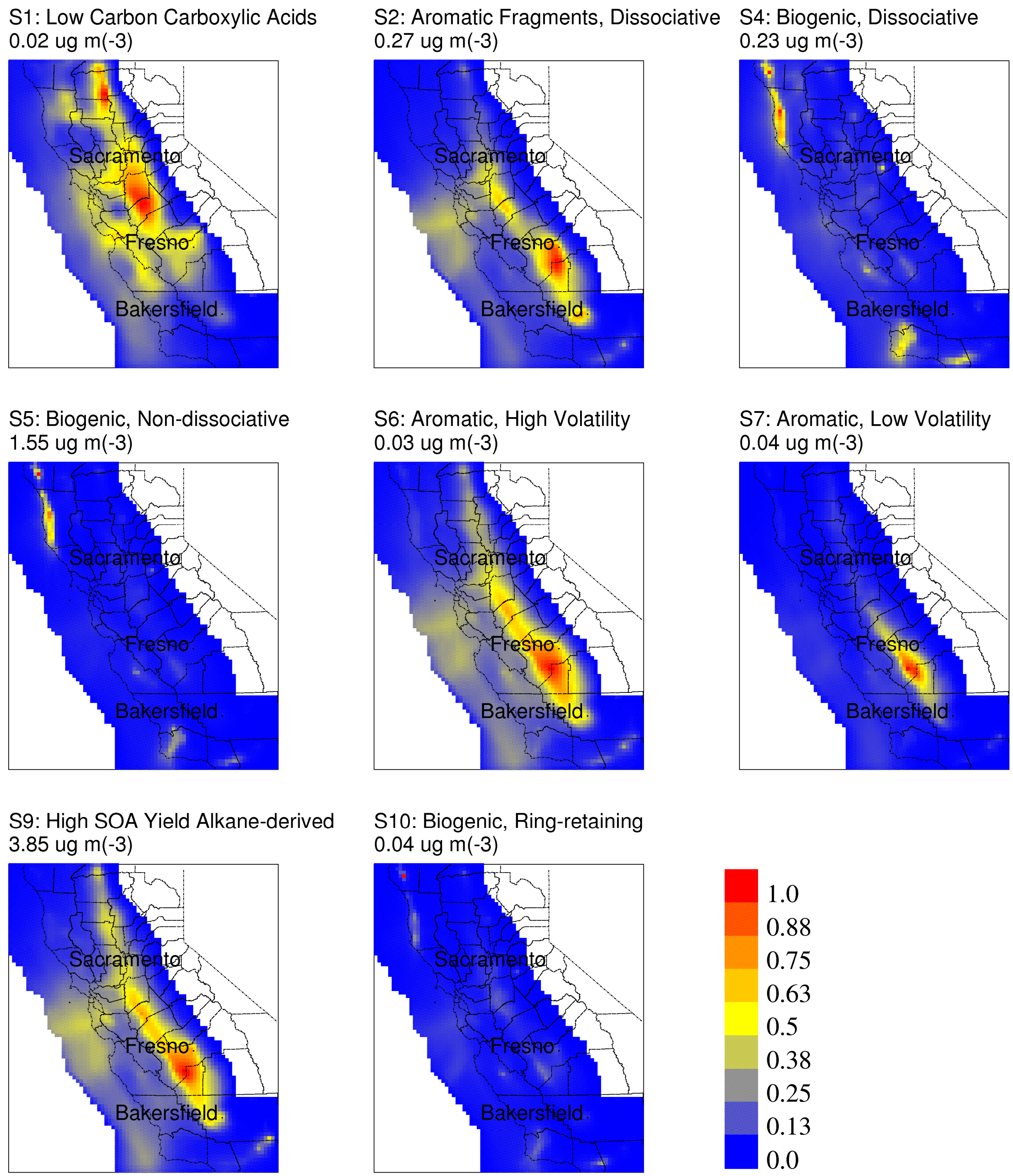


c.) Bakersfield

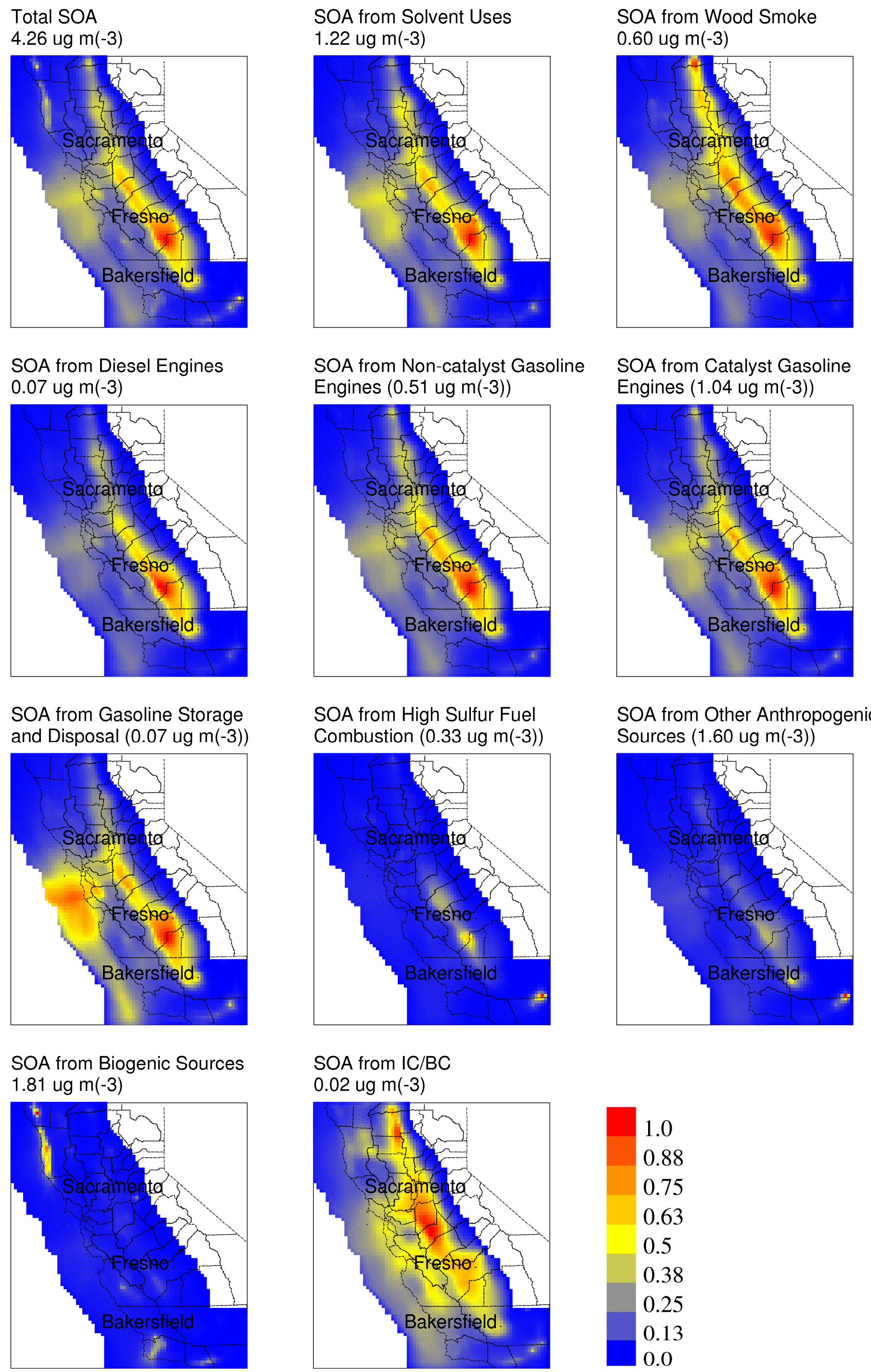
OA Fractional Bias Using CACM



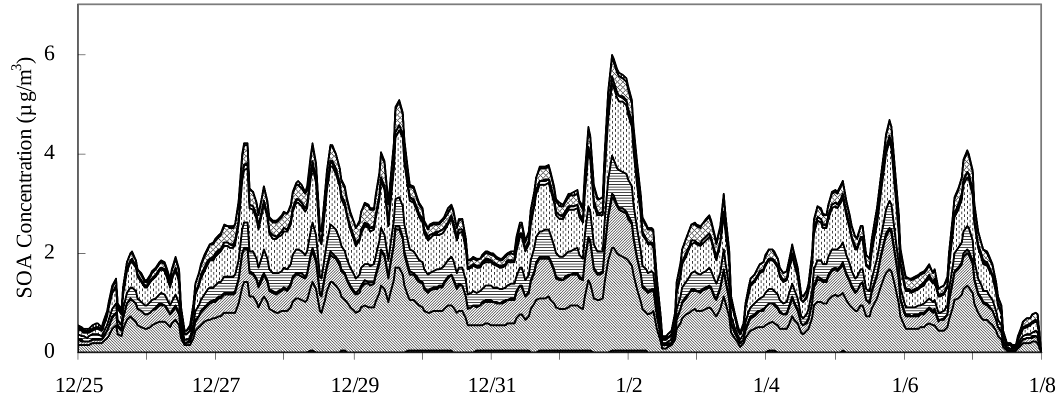
SOA contributions from selected chemical surrogate species



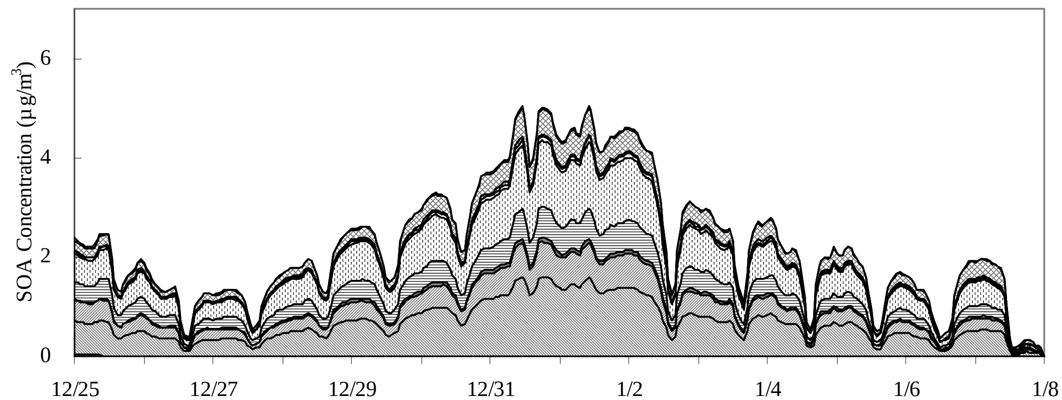
Predicted SOA Source Contributions Using CACM December 25, 2000 and January 7, 2001.



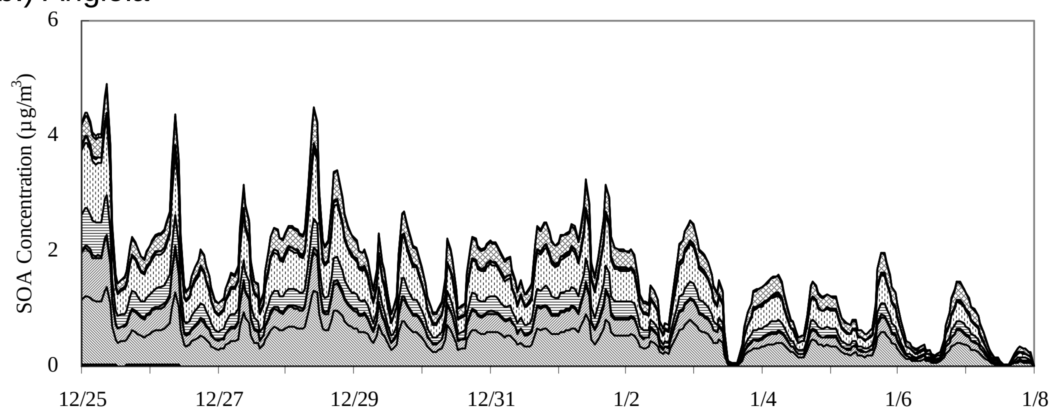
SOA Source Contributions



a.) Fresno



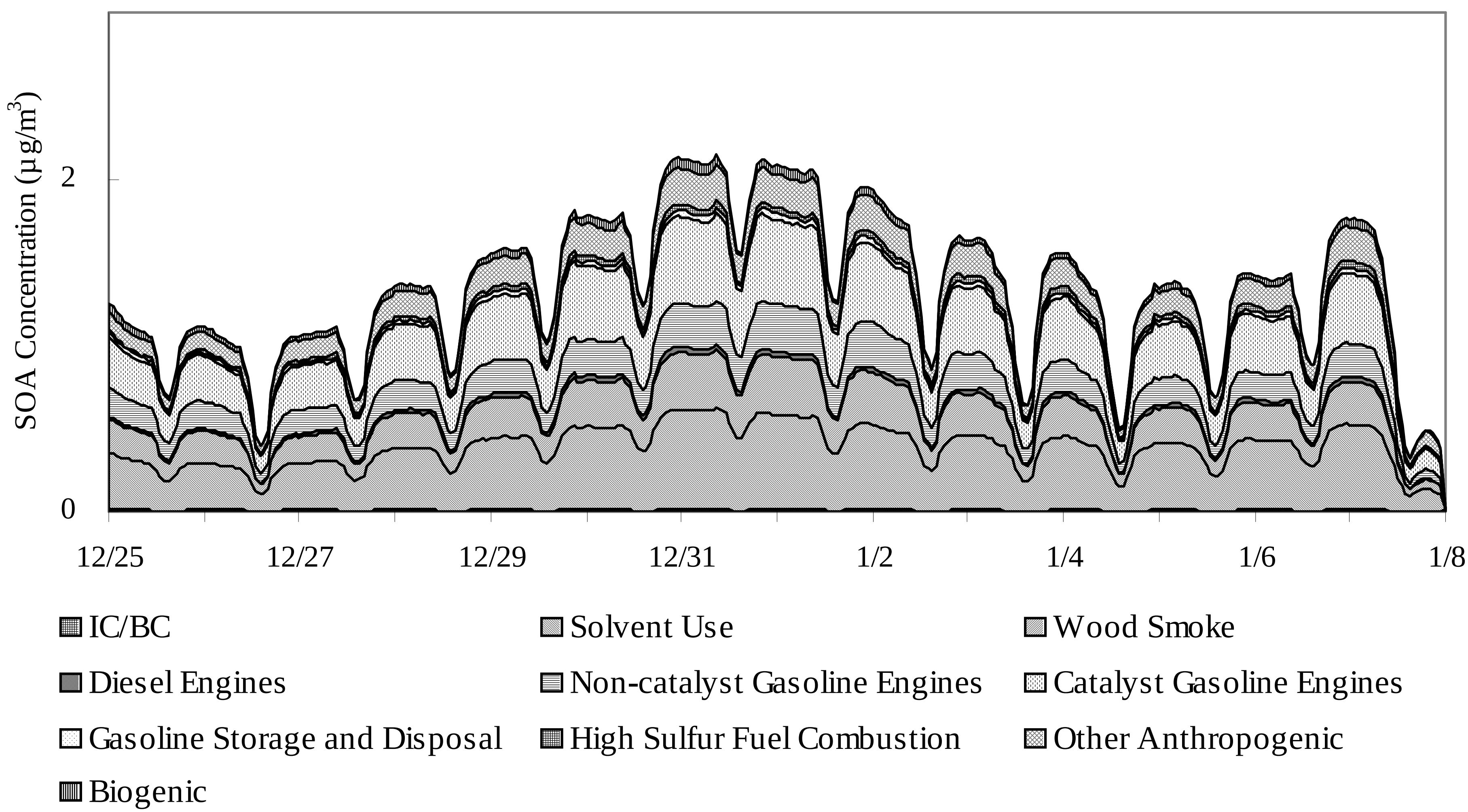
b.) Angiola



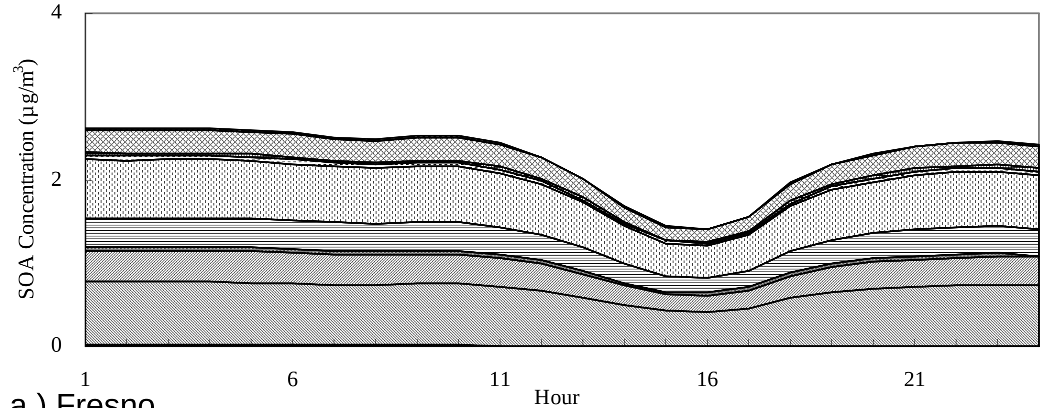
c.) Bakersfied

IC/BC
Wood Smoke
Non-catalyst Gasoline Engines
Gasoline Storage and Disposal
Other Anthropogenic
Solvent Use
Diesel Engines
Gasoline Engines
High Sulfur Fuel Combustion
Biogenic

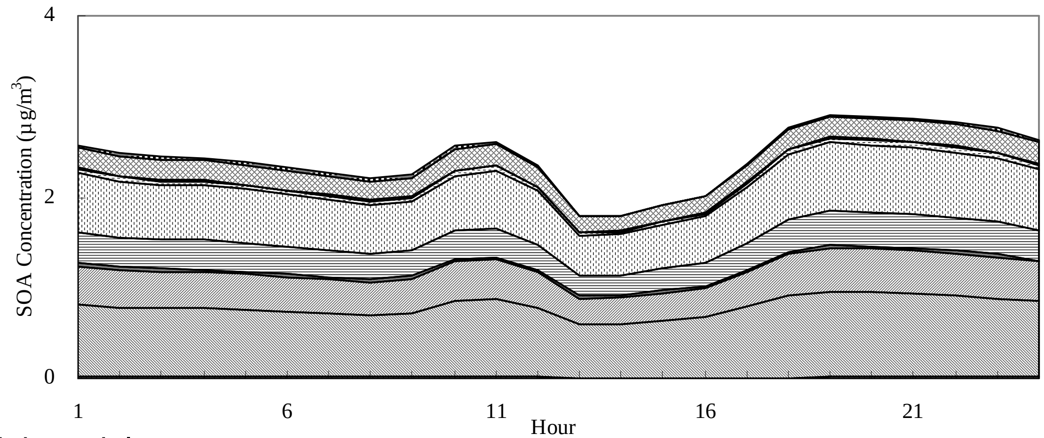
SJV Average SOA Source Contributions



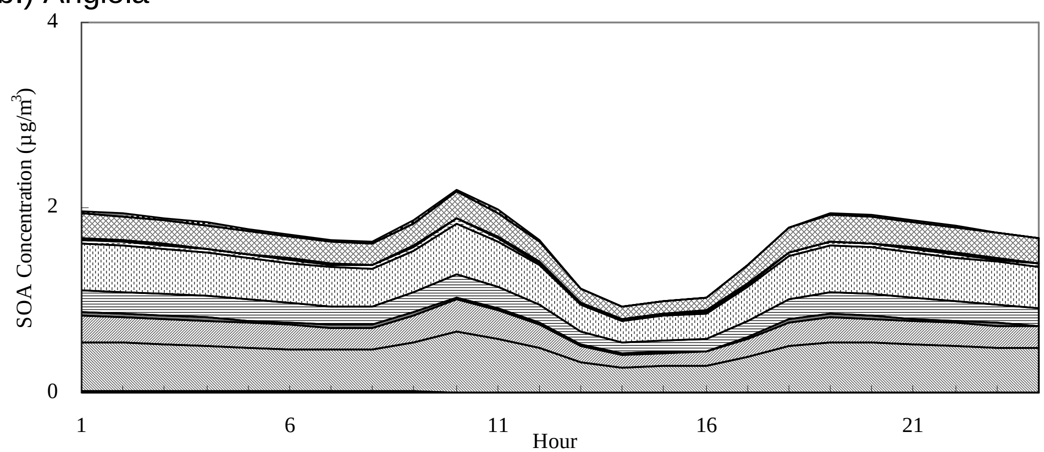
SOA Diurnal Variation December 25, 2000 and January 7, 2001.



a.) Fresno



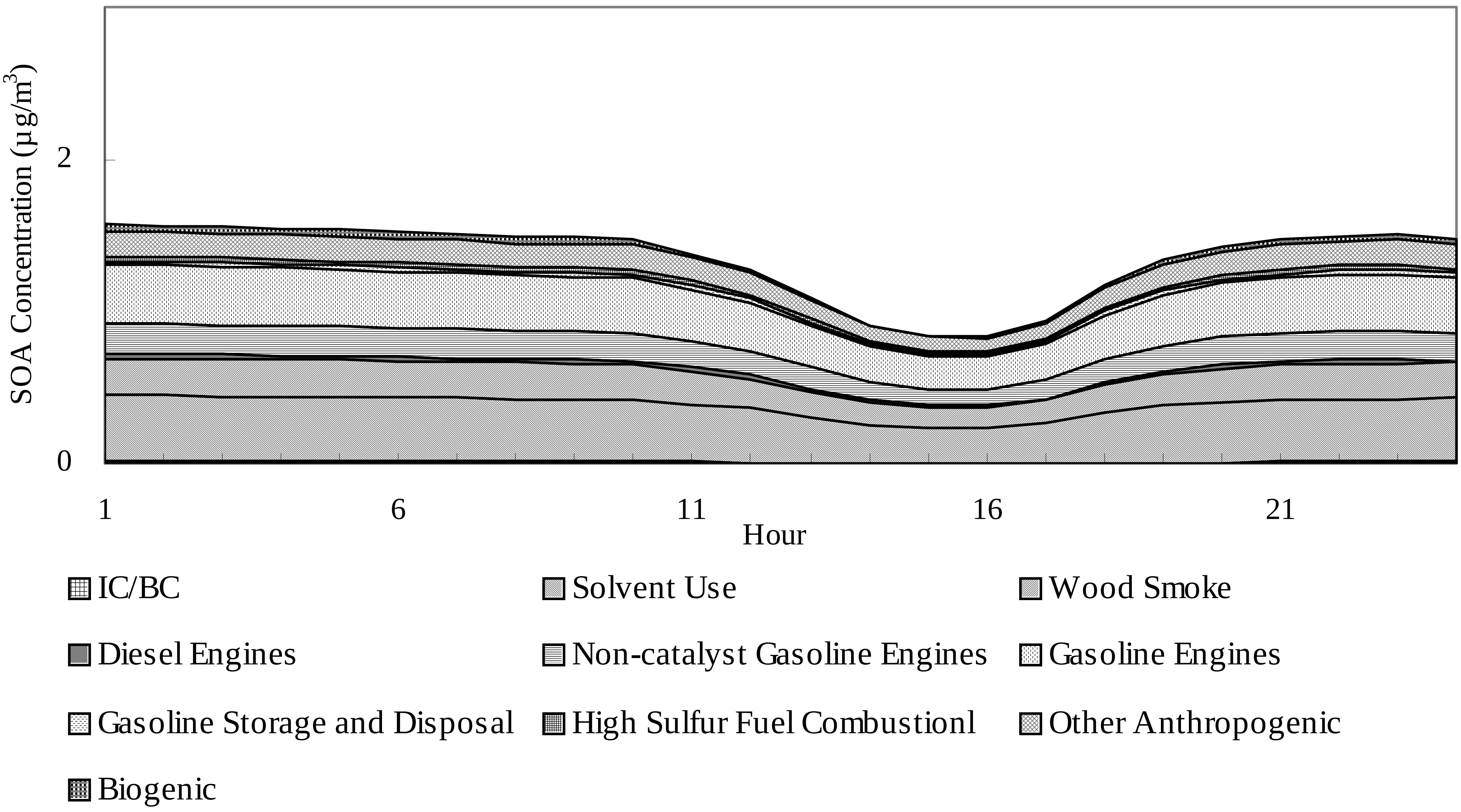
b.) Angiola



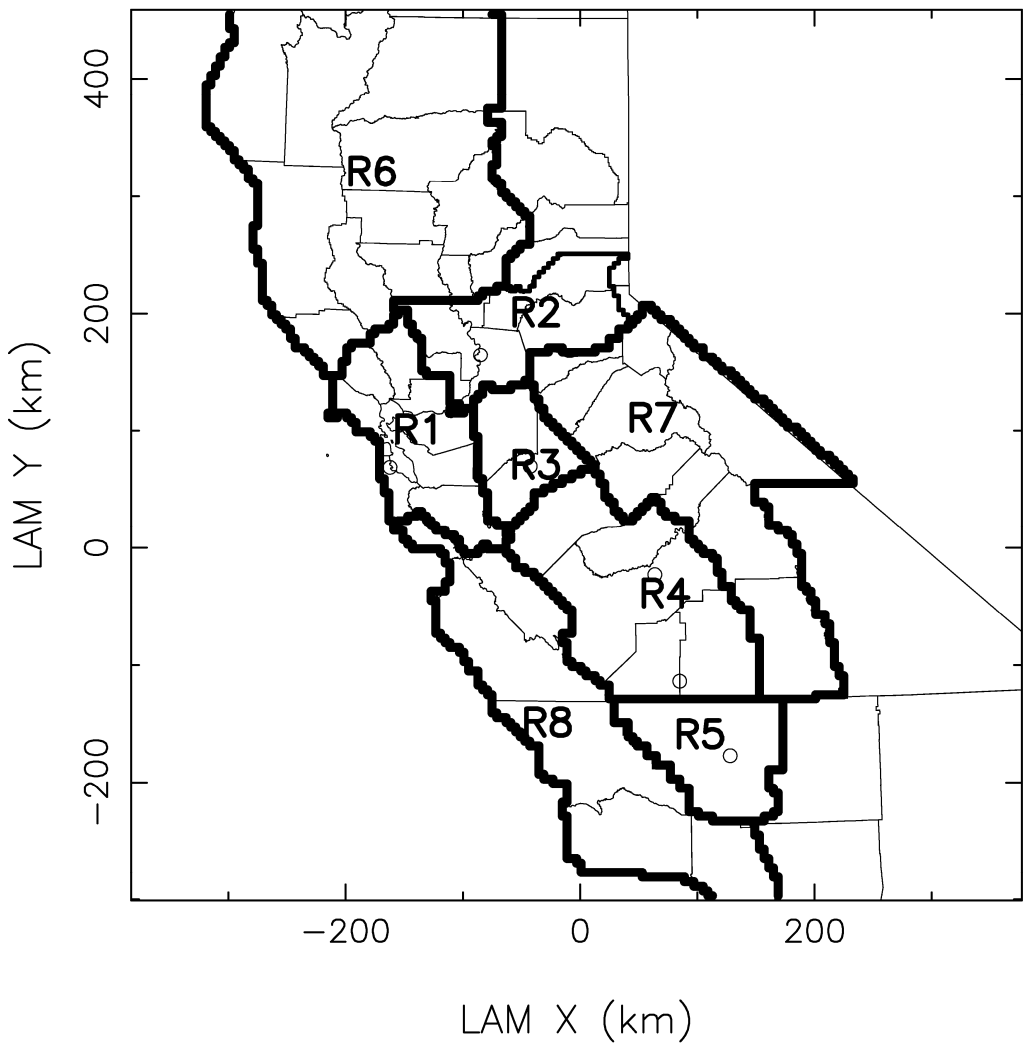
c.) Bakersfield

IC/BC
Solvent Use
Wood Smoke
Diesel Engines
Non-catalyst Gasoline Engines
Gasoline Engines
Gasoline Storage and Disposal
High Sulfur Fuel Combustion
Other Anthropogenic
Biogenic

SJV Average SOA Diurnal Variation

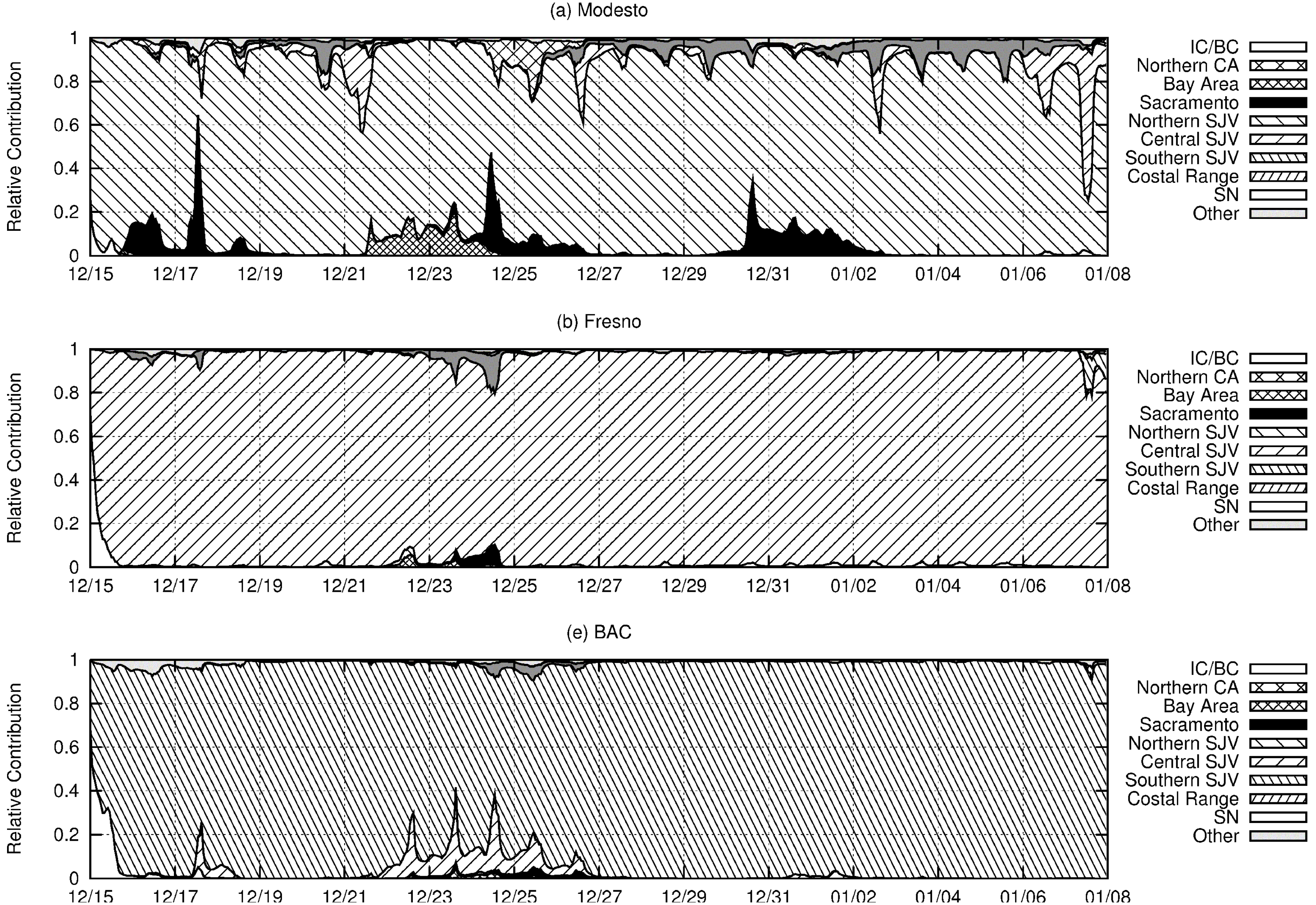


How Much PM Does Each Region Contribute to Other Regions?



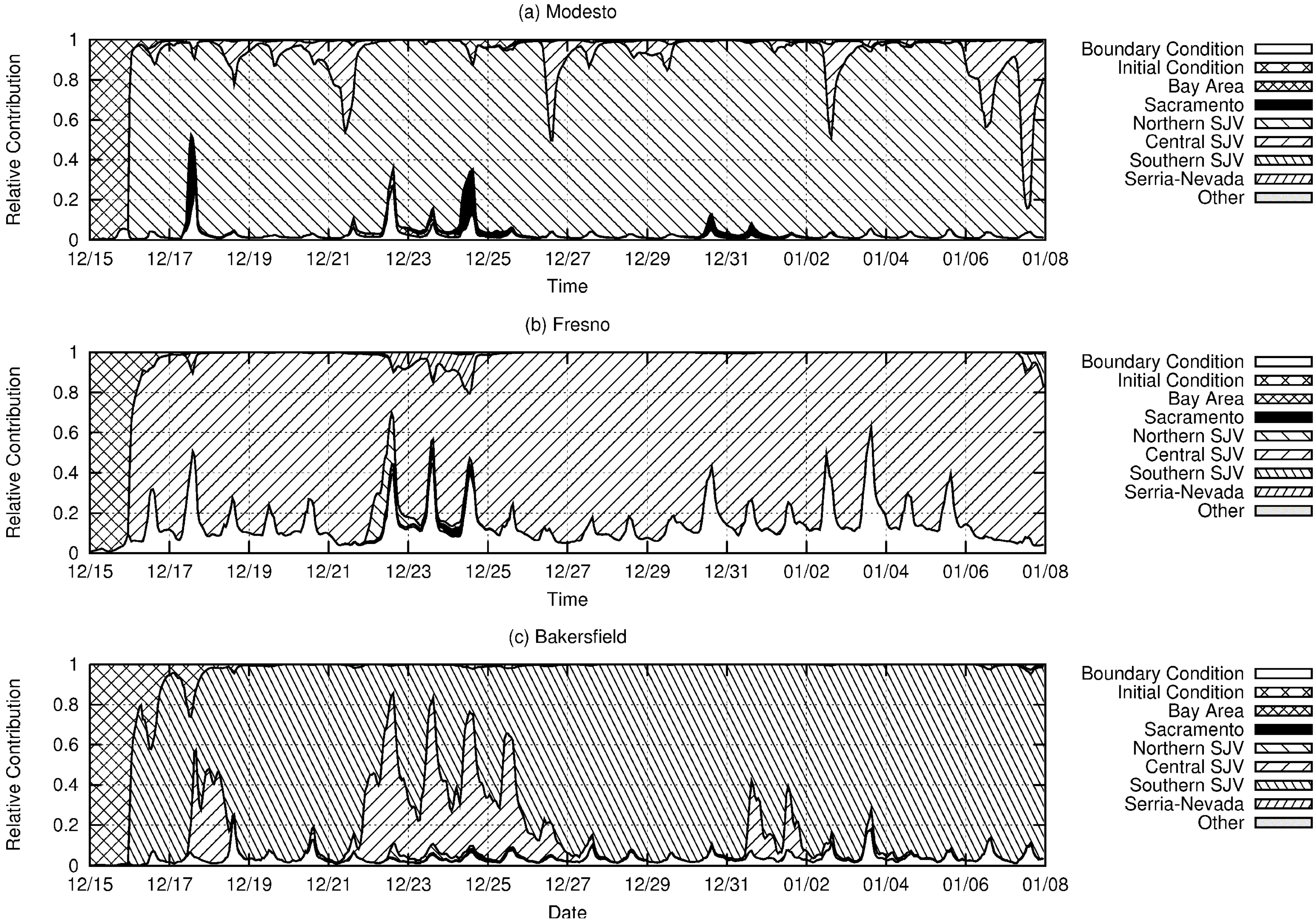
Source: Q. Ying, and M. Kleeman "Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode", Atmos. Env., submitted for publication, 2008.

OC Region Contributions



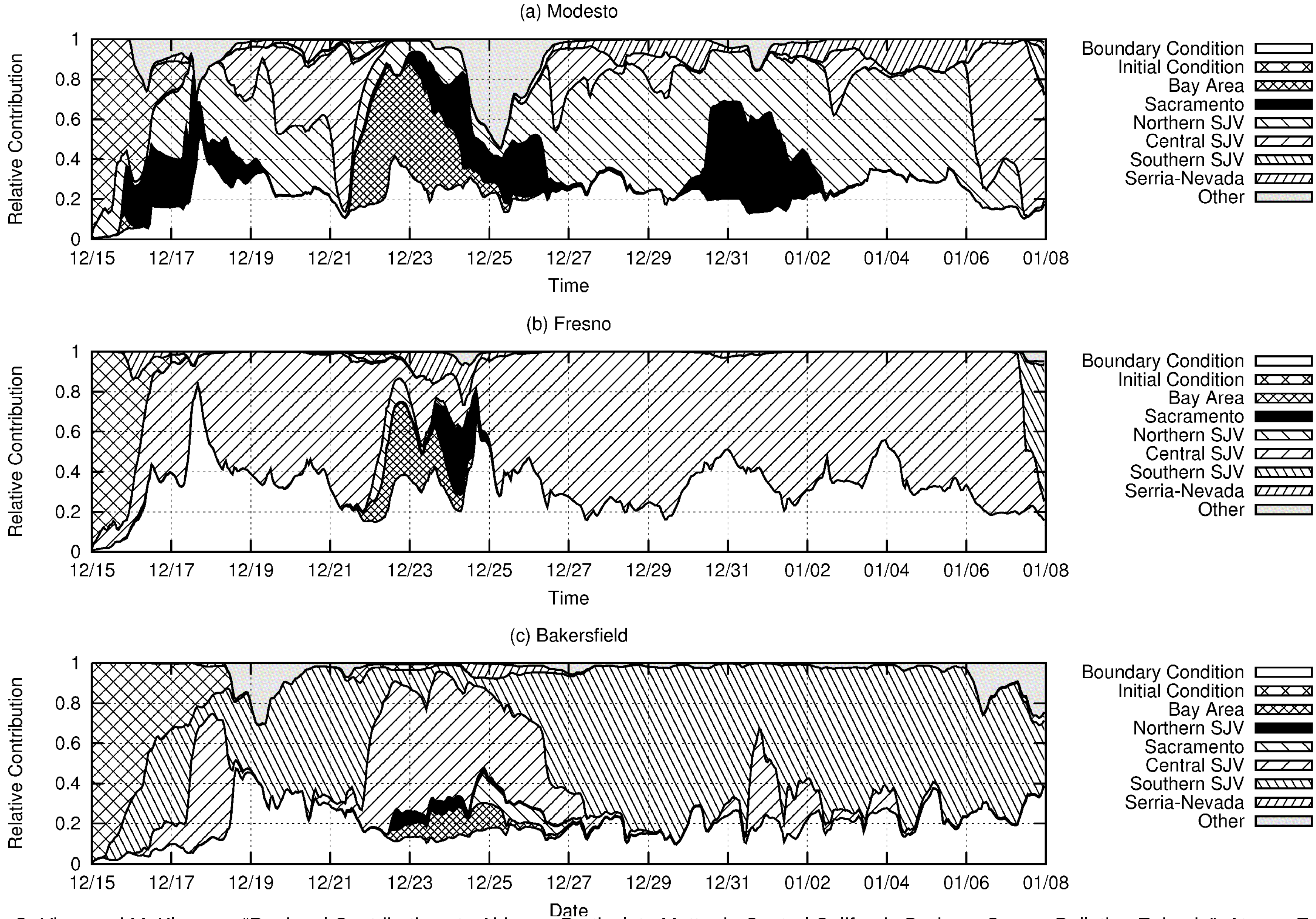
Source: Q. Ying, and M. Kleeman "Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode", Atmos. Env., submitted for publication, 2008.

NH_4^+ Region Contributions



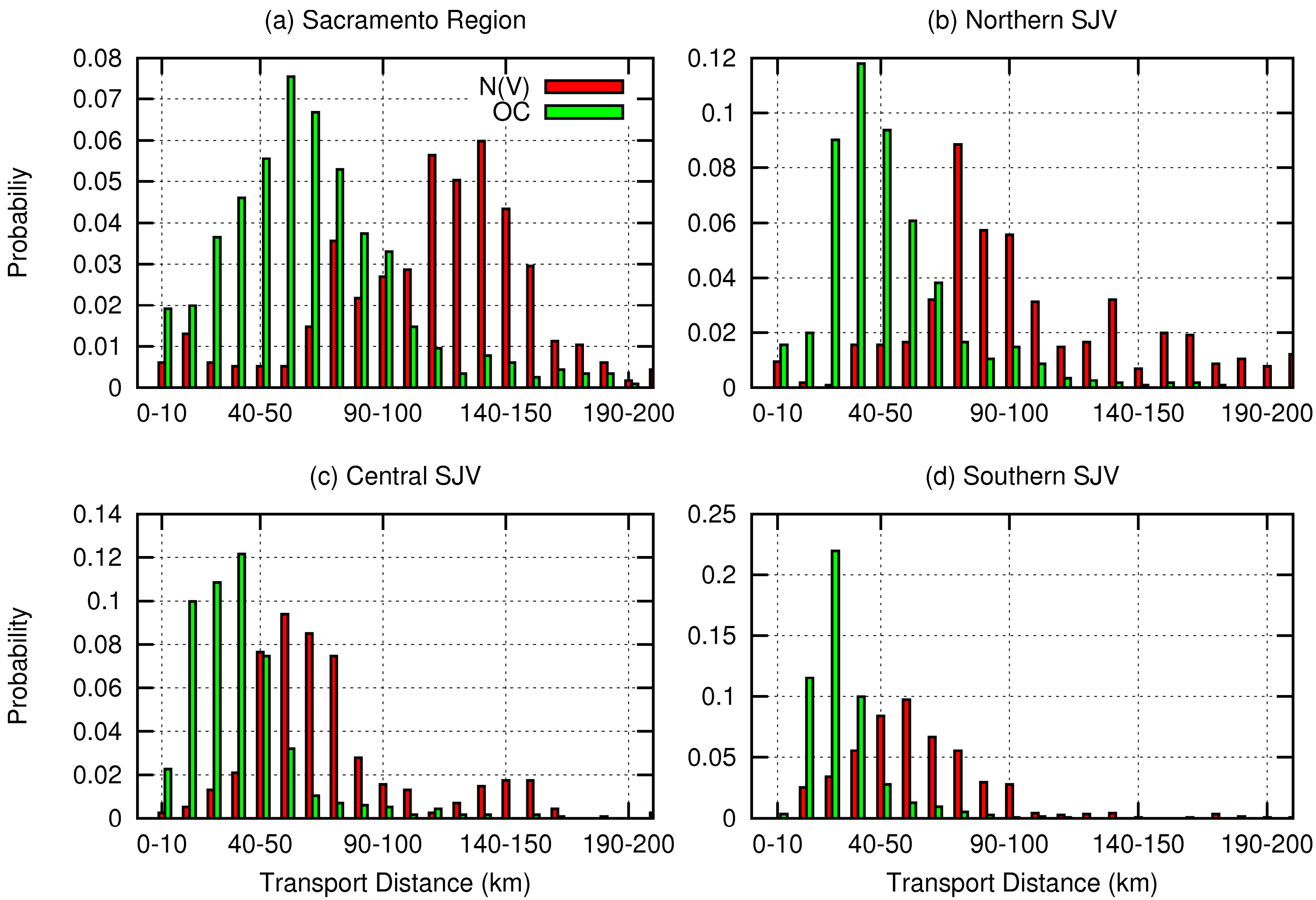
Source: Q. Ying, and M. Kleeman "Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode", Atmos. Env., submitted for publication, 2008.

NO_3^- Region Contributions



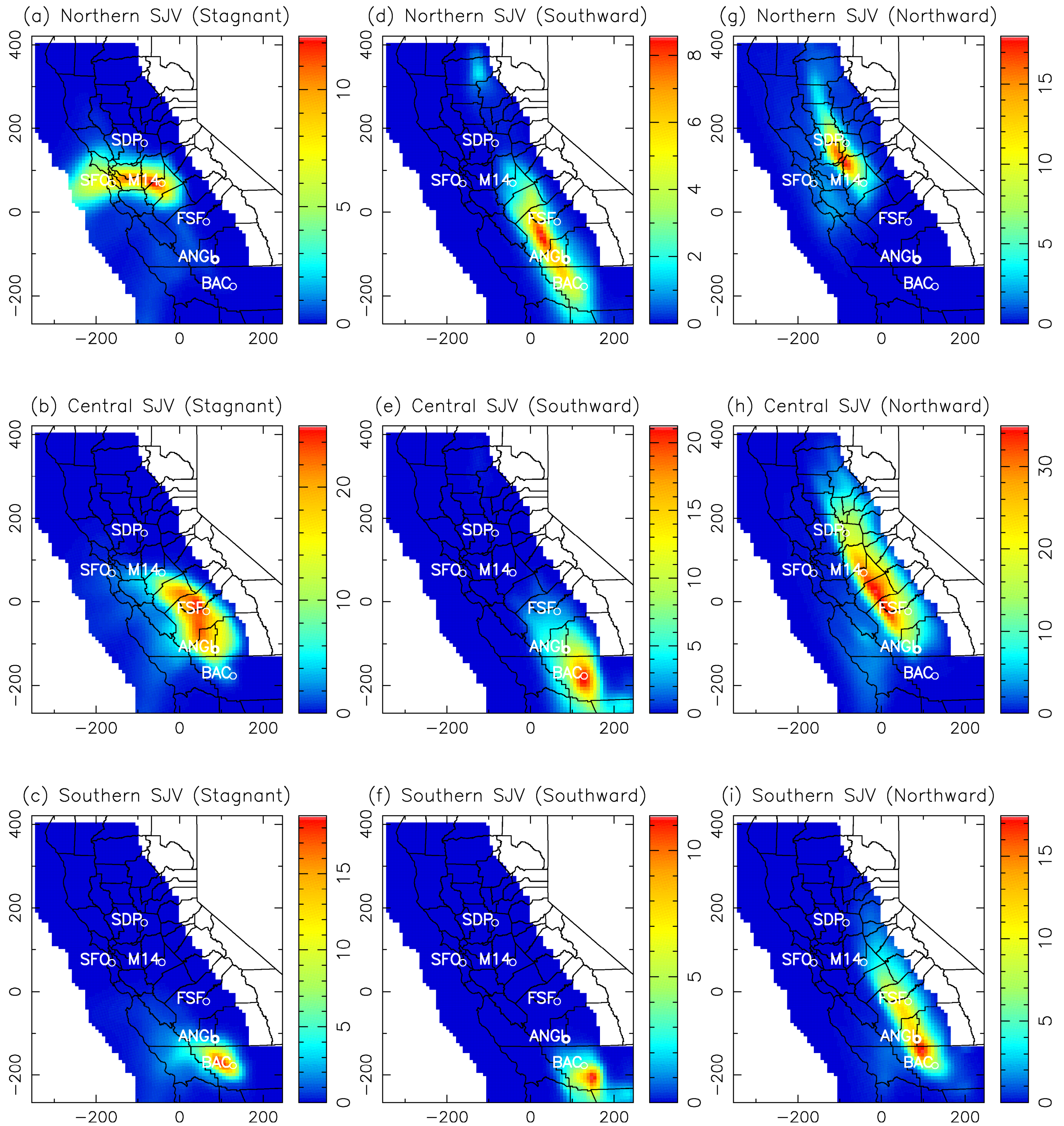
Source: Q. Ying, and M. Kleeman "Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode", Atmos. Env., submitted for publication, 2008.

Distribution of Transport Distances



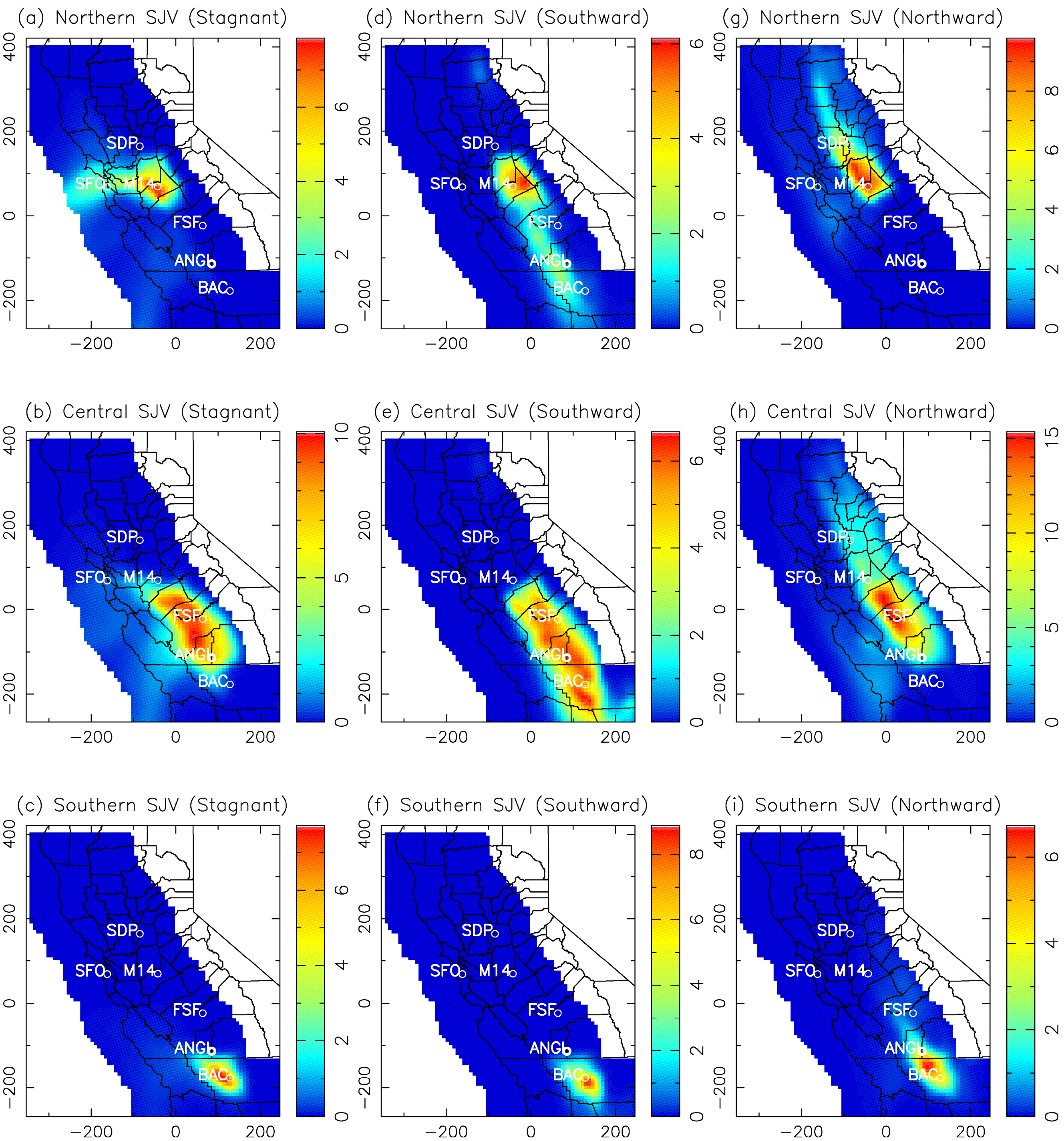
Source: Q. Ying, and M. Kleeman “Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode”, Atmos. Env., submitted for publication, 2008.

PM2.5 NO3-



Source: Q. Ying, and M. Kleeman "Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode", Atmos. Env., submitted for publication, 2008.

PM2.5 NH4+



Source: Q. Ying, and M. Kleeman "Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode", Atmos. Env., submitted for publication, 2008.

Regional Contribution Summary

Table 1

24-hour averaged $PM_{2.5}$ nitrate contribution from each source sub-region (Sx) to other receptor sub-regions (Rx). Figure 1 shows region designations: 0=boundary conditions; 1=Bay Area; 2=Sacramento; 3=northern SJV; 4=central SJV; 5=southern SJV; 6=northern Sacramento Valley; 7=Sierra Mountains; 8=Other.

	S0	S1	S2	S3	S4	S5	S6	S7	S8	SUM
R1	2.1%	1.5%	1.8%	1.4%	0.4%	0.0%	0.2%	0.2%	0.7%	8.2%
R2	1.1%	0.2%	1.3%	0.3%	0.3%	0.0%	0.1%	0.1%	0.8%	4.3%
R3	1.3%	0.3%	0.8%	1.7%	1.0%	0.0%	0.1%	0.4%	0.2%	5.9%
R4	5.0%	0.8%	0.9%	1.7%	17.7%	0.7%	0.8%	0.4%	0.3%	28.3%
R5	1.4%	0.3%	0.2%	0.2%	3.1%	3.1%	0.4%	0.0%	0.2%	9.0%
R6	3.4%	1.1%	0.6%	1.1%	4.0%	1.4%	2.0%	0.1%	0.7%	14.5%
R7	0.5%	0.1%	0.1%	0.1%	0.3%	0.0%	0.1%	0.2%	0.0%	1.5%
R8	8.4%	3.9%	2.3%	1.7%	2.4%	0.6%	2.3%	0.2%	6.5%	28.4%
SUM	23.2%	8.2%	8.1%	8.2%	29.3%	6.0%	6.0%	1.6%	9.4%	100.0%

Source: Q. Ying, and M. Kleeman "Regional Contributions to Airborne Particulate Matter in Central California During a Severe Pollution Episode", Atmos. Env., submitted for publication, 2008.

Conclusions

- Basecase model performance using diagnostic meteorology captures major pollutant trends
- Mechanistic primary source apportionment (grid model) agrees well with CMB results
- Source contributions to primary PM
  - Wood smoke at urban locations
- Source contributions to secondary nitrate
  - Diesel engines contribute most of the NOx
  - Gasoline engines also significant

Conclusions

- Primary and secondary PM concentrations are dominated by local sources
- Transport distance for Nitrate > Ammonium Ion > Organic Carbon
- Transport between regions seems relatively low during CRPAQS stagnant conditions
  - Material mixed to upper layers is quickly transported out of the domain.

Conclusions

- SOA can account for a significant fraction of the OC at rural locations
  - Solvent use is a dominant source

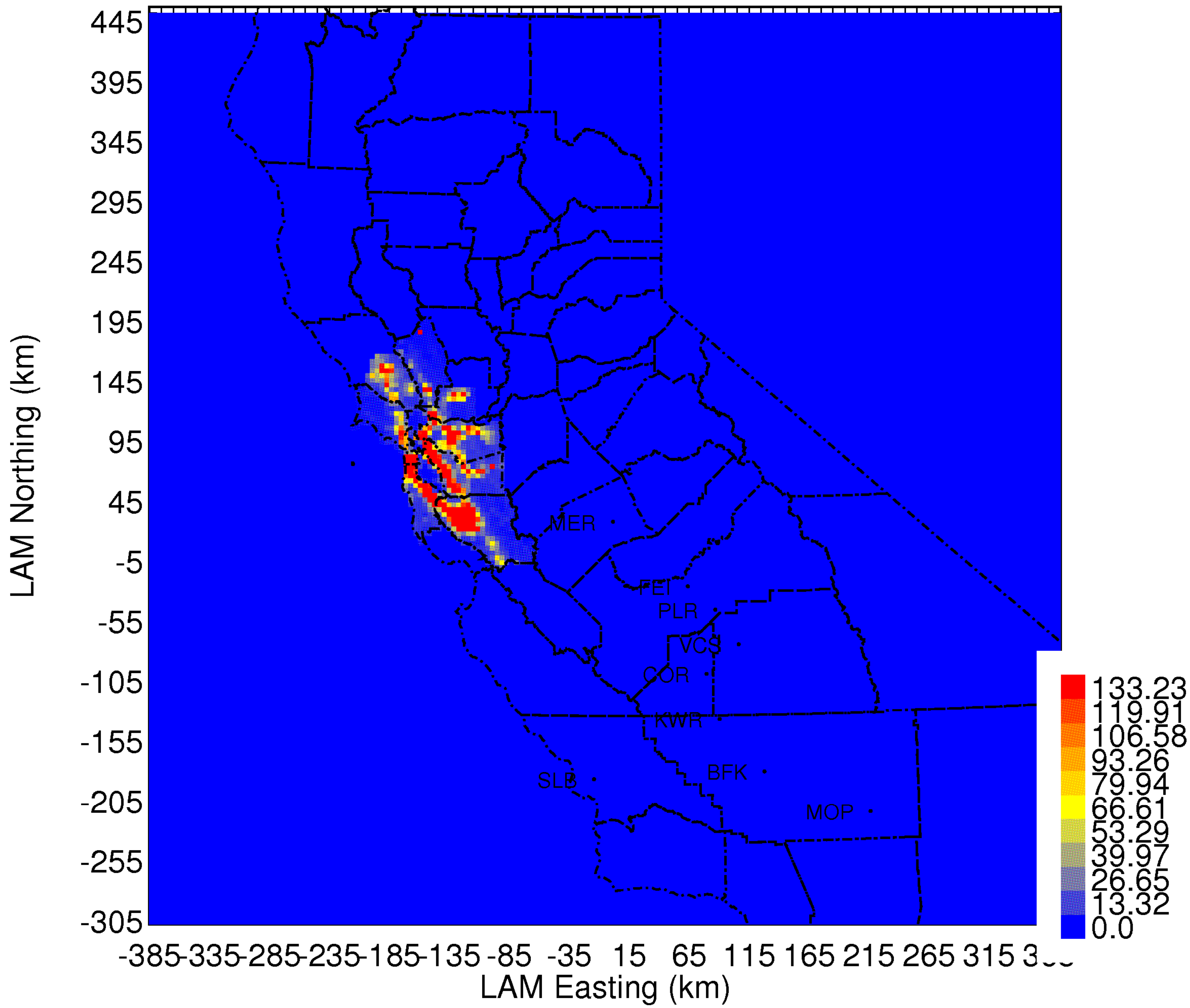
Future Work

- Visibility Source Apportionment
- Compare particle mixing state and composition to ATOFMS single particle measurements
  - Requires that we simulate the Feb episode (IOP4) because this is the only useful ATOFMS data

Supporting Slides

Secondary Organic Aerosol Mechanism (SOAM)

- SAPRC90 mechanism with enhancements to describe formation of condensable organics
  - 126 species
  - 208 reactions
- Modifications for source apportionment of NO_3^-, SO_4^{2-}, and NH_4^+
  - 298 species
  - 1261 reactions



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LAM Northing (km)
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LAM Easting (km)
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COR
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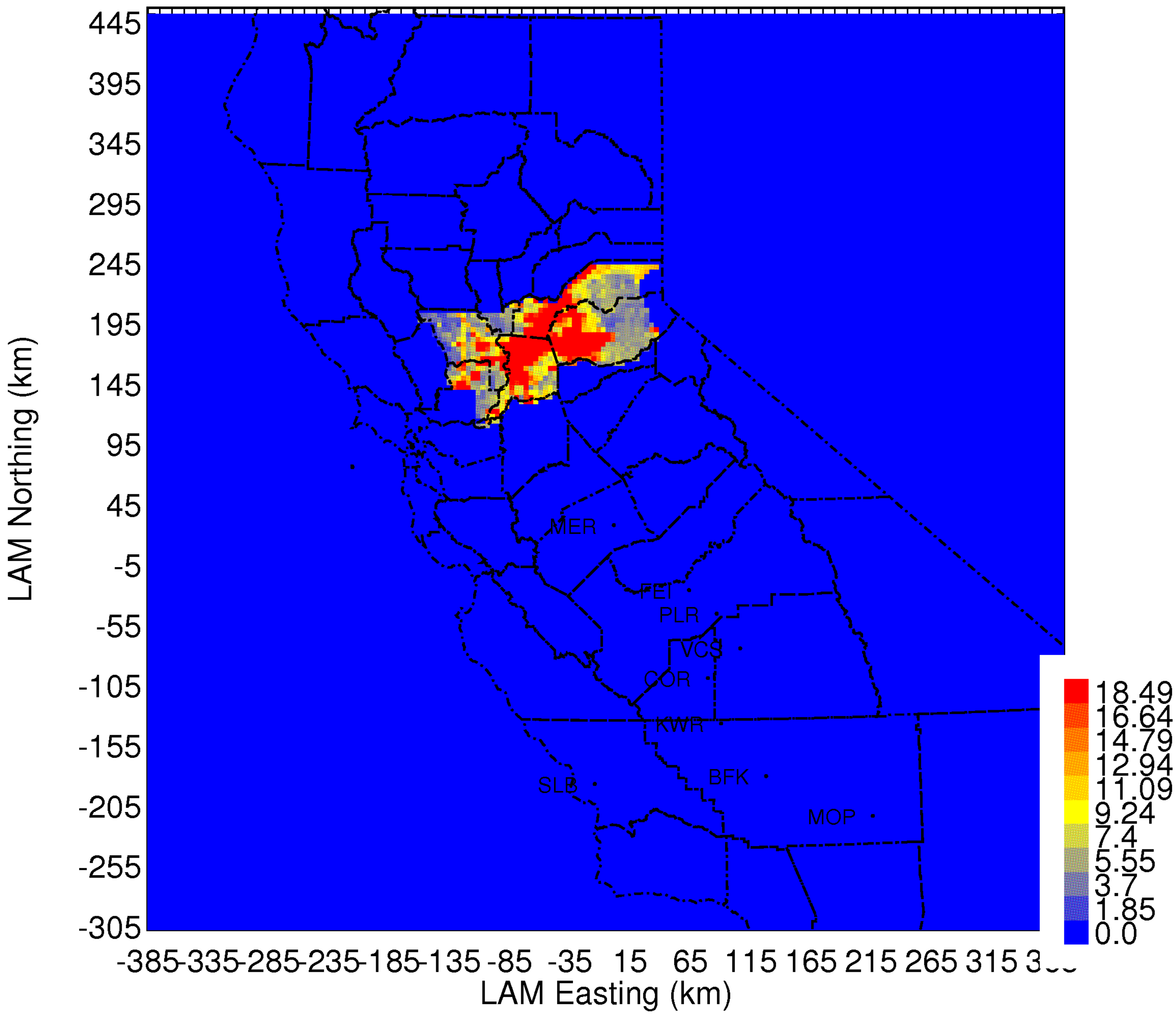

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LAM Northing (km)
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LAM Easting (km)
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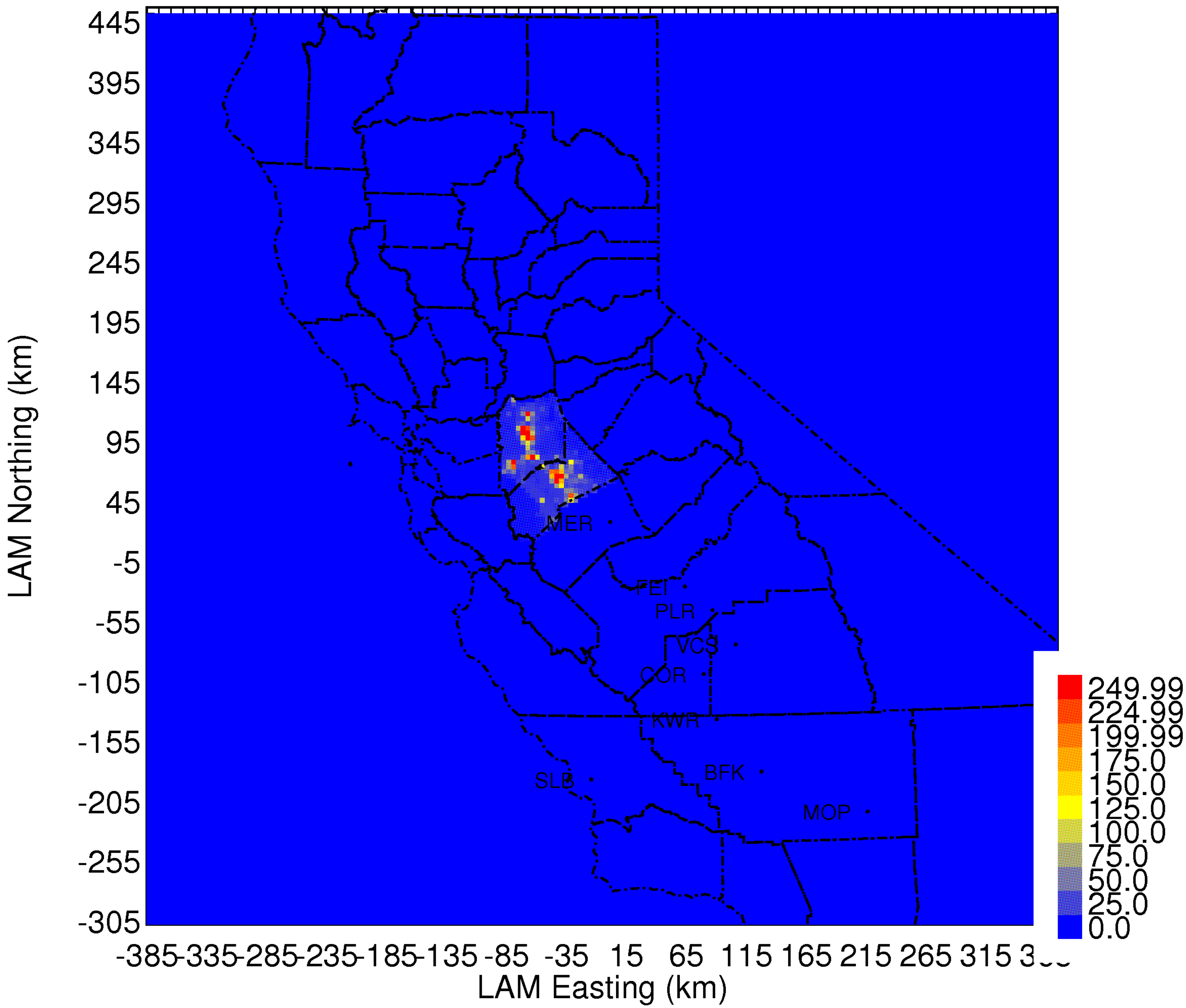

LAM Northing (km)
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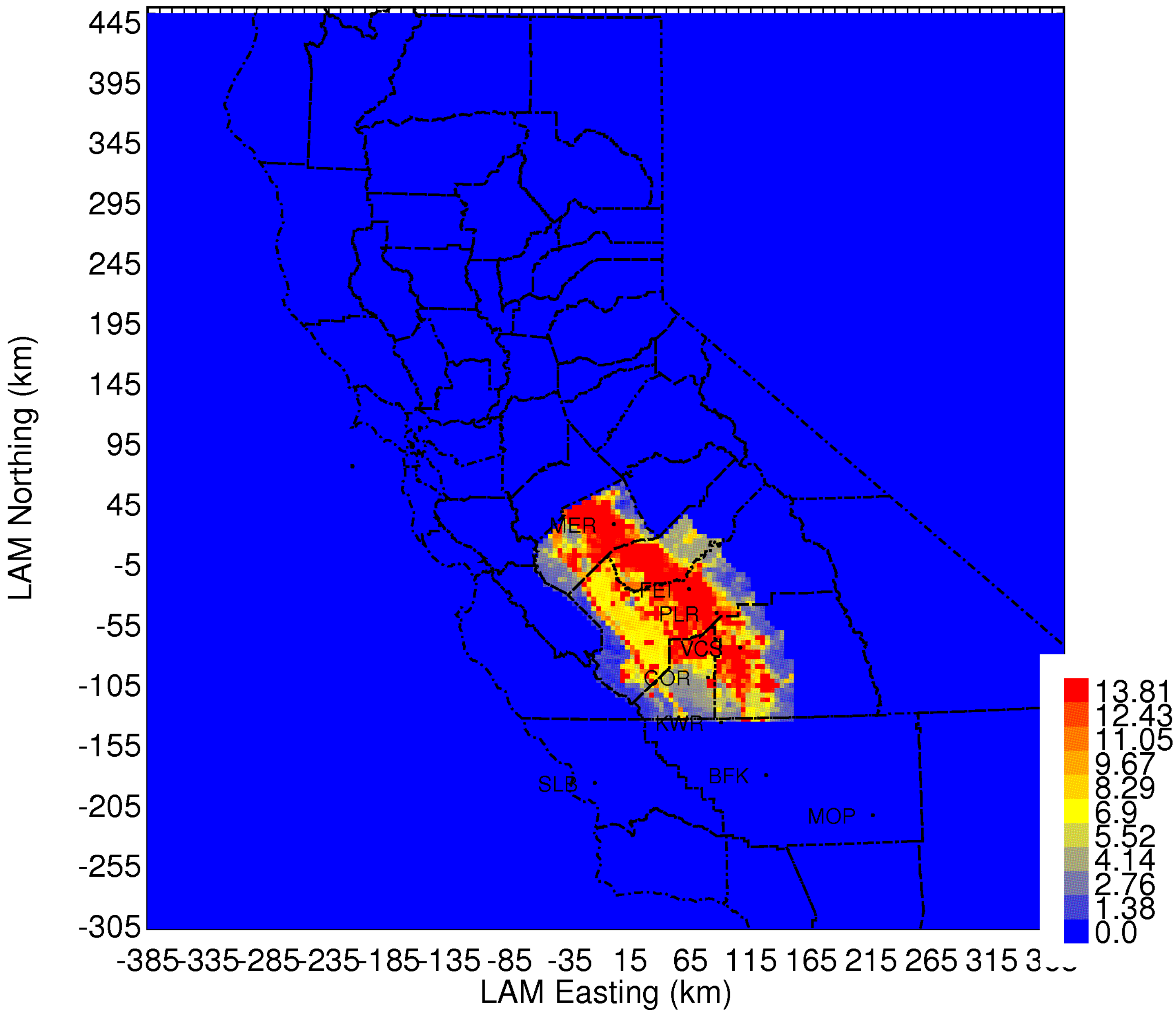
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LAM Easting (km)
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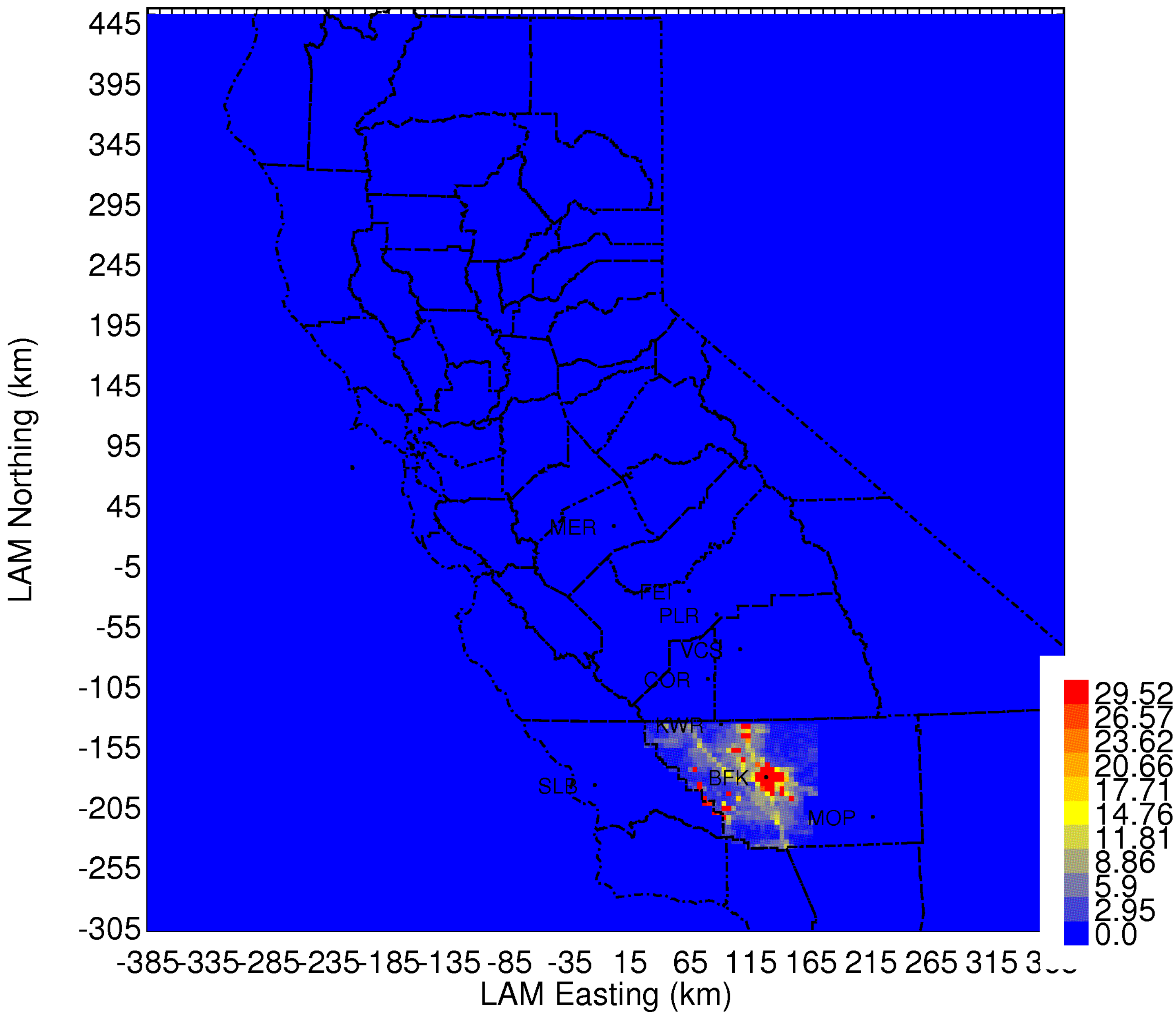

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LAM Easting (km)
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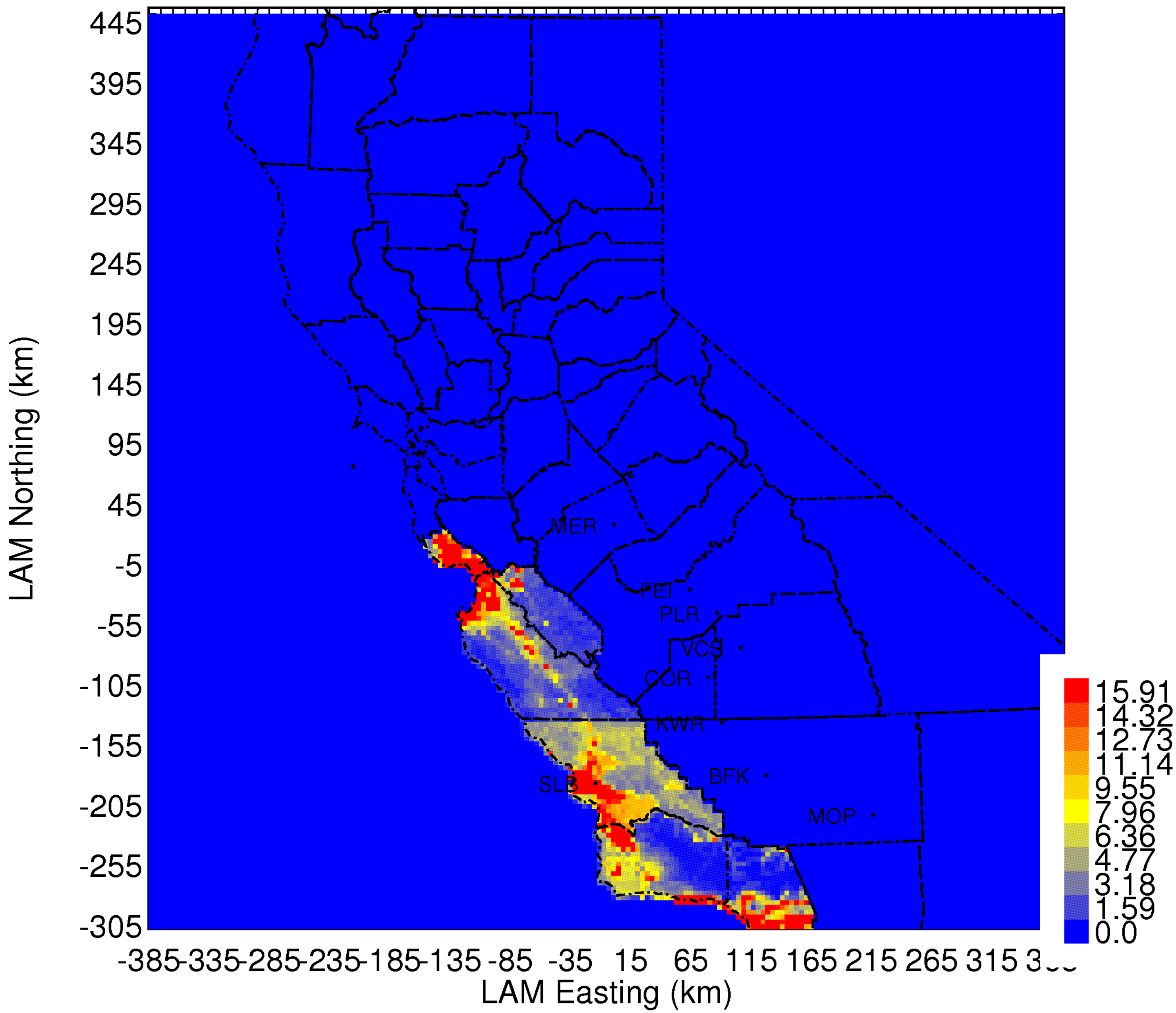
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LAM Northing (km)
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LAM Easting (km)
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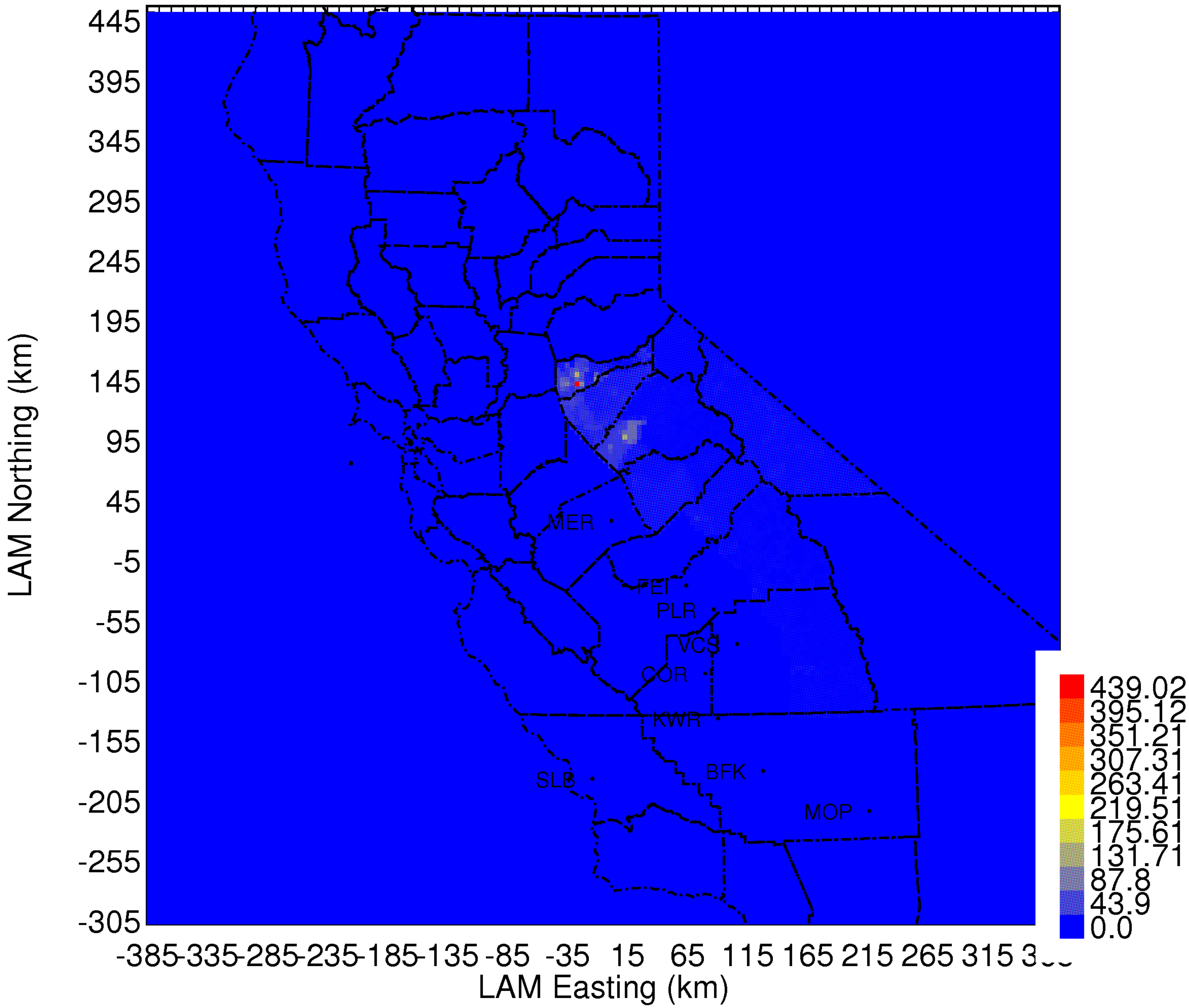

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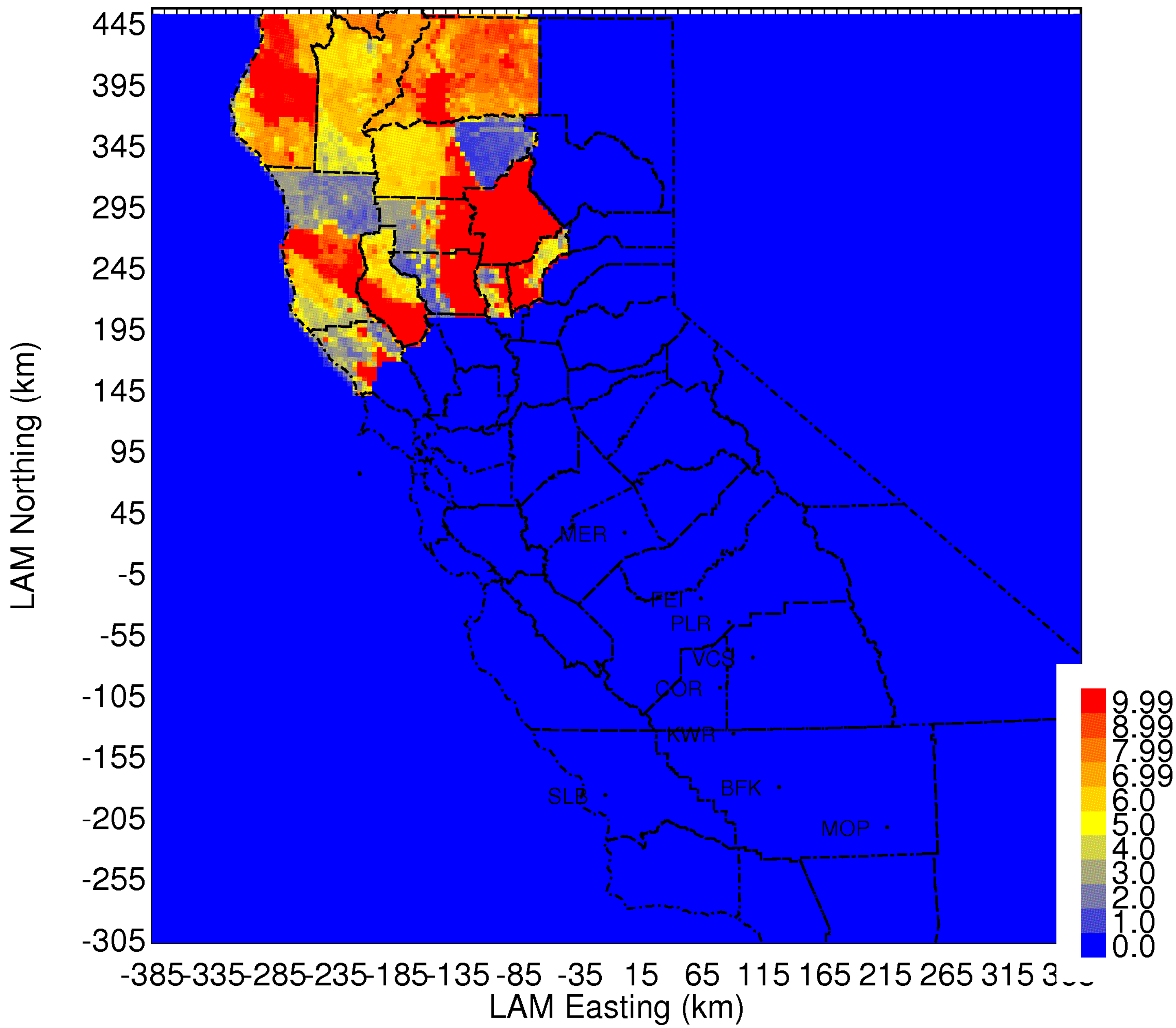
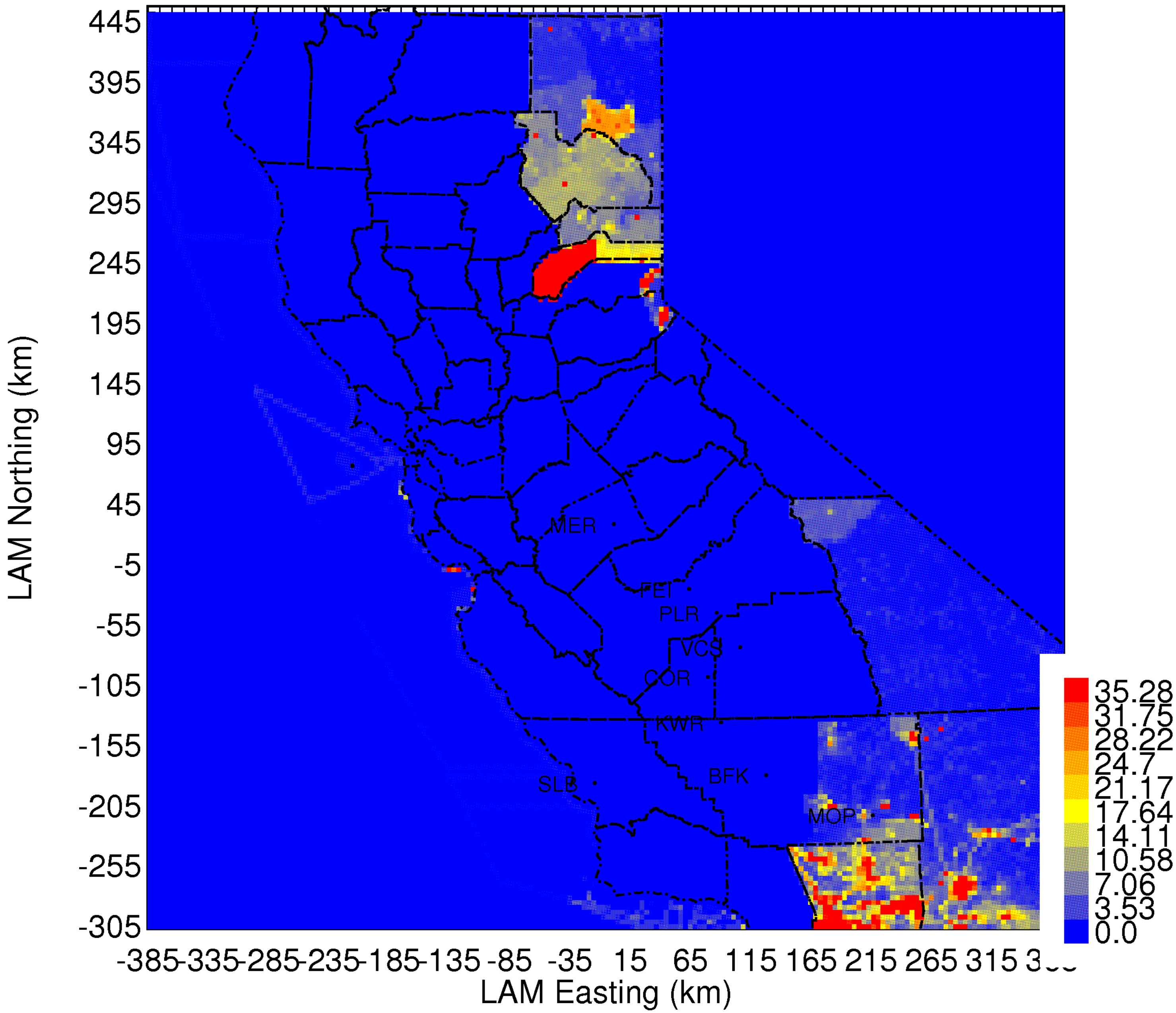
43.9
0.0


LAM Northing (km)
445
395
345
295
245
195
145
95
45
-5
-55
-105
-155
-205
-255
-305
-385-335-285-235-185-135 -85 -35 15 65 115 165 215 265 315
LAM Easting (km)
MER
FEI
PLR
VCS
COR
KWR
SLB
BFK
MOP
9.99
8.99
7.99
6.99
6.0
5.0
4.0
3.0
2.0
1.0
0.0



LAM Northing (km)
LAM Easting (km)
445
395
345
295
245
195
145
95
45
-5
-55
-105
-155
-205
-255
-305
-385
-335
-285
-235
-185
-135
-85
-35
15
65
115
165
215
265
315
MER
FEI
PLR
VCS
COR
KWR
BFK
SLB
MOP
35.28
31.75
28.22
24.7
21.17
17.64
14.11
10.58
7.06
3.53
0.0